

## 2. SITE HISTORICAL OVERVIEW

### 2.1 Site Description

The PORTS facility is one of two operating uranium enrichment production facilities in the United States. Both facilities are owned by the DOE. The other operating facility is located in Paducah, Kentucky. (A third uranium enrichment facility, in Oak Ridge, Tennessee, was placed in standby in 1985 and shut down in 1987). Each facility utilizes the gaseous diffusion process to enrich uranium from a natural state of less than 1%  $^{235}\text{U}$  to increased concentrations varying from 2 to 5%  $^{235}\text{U}$  for use as fuel for nuclear power generation. The Paducah facility presently enriches uranium to approximately 2.5%  $^{235}\text{U}$  and then ships it to PORTS for further enrichment. PORTS had the capability of achieving a higher percentage of enrichment; however, highly enriched uranium operations were shut down beginning in 1991.



PORTSMOUTH PLANT Circa 1958

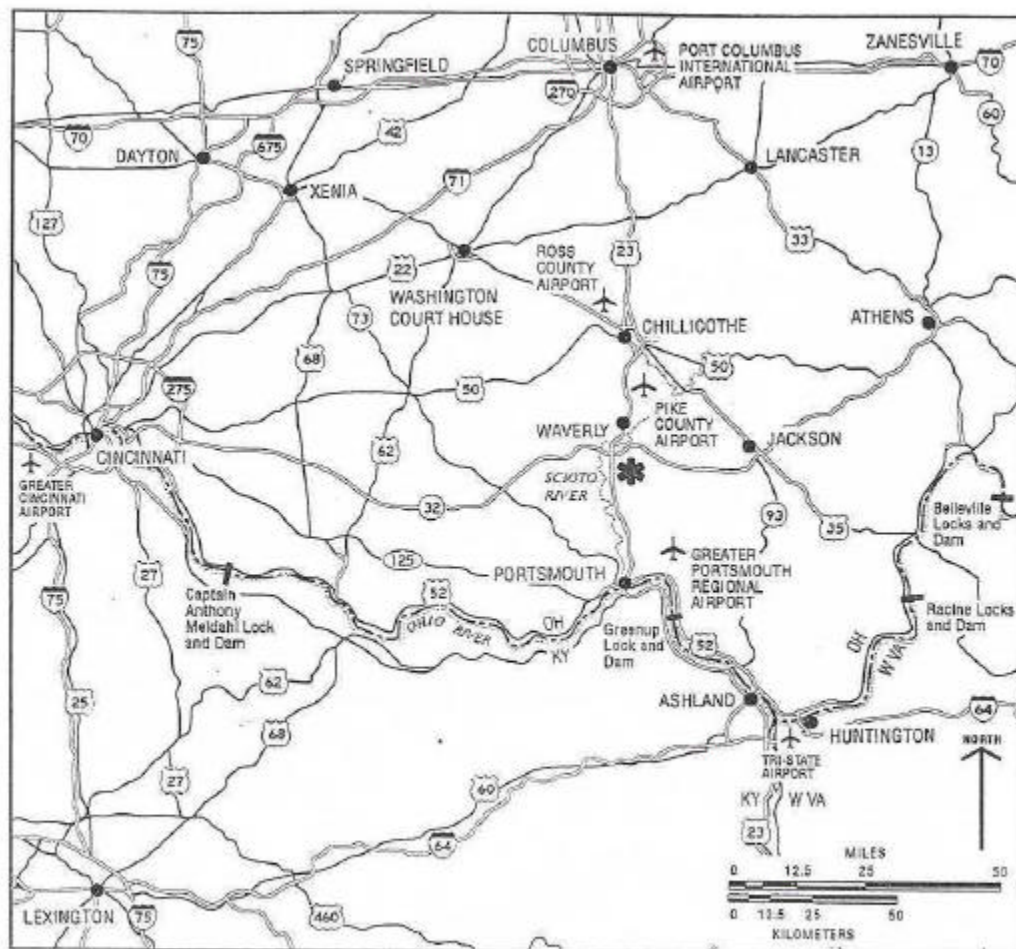
In June 1951, the Atomic Energy Commission (AEC) began design studies that would lead to the construction of a gaseous diffusion plant that could be added to the Oak Ridge—Paducah complex and provide  $^{235}\text{U}$  production at rates substantially above those of the existing complex. The AEC began site selection for a new gaseous diffusion plant and selected PORTS in 1952.

PORTS consists of 109 buildings occupying 500 acres located on a 3708 acre DOE-owned reservation in Pike County in south central Ohio about two miles east of the Scioto River and 18 miles north of Portsmouth, Ohio (refer to regional location, Figure 2.1-1). Construction of the plant began in late 1952 and production of enriched uranium began in early FY 1955, one year before completion of construction. In the late 1970's, PORTS was chosen as the site for construction of a new uranium enrichment facility utilizing gas centrifuge technology. Construction of the Gas Centrifuge Enrichment Plant (GCEP) began in 1979 in an area southwest of and adjacent to the existing gaseous diffusion plant. Construction of this facility was halted in the summer of 1985.

With the Energy Policy Act of 1992, the responsibility for the operation of the gaseous diffusion plant at PORTS transferred to the newly created USEC effective July 1, 1993. With this transfer of responsibility, DOE leased to USEC property shown in Portsmouth Gaseous Diffusion Plant Building Lease Status, Figure 2.1-2. Although the USEC has managed the gaseous diffusion operations at PORTS since July 1, 1993, the DOE continues to have a significant presence, particularly in the area of environmental restoration and the responsibility for treating and disposing of wastes resulting from GDP operations prior to July 1, 1993.

**Figure 2.1-1**

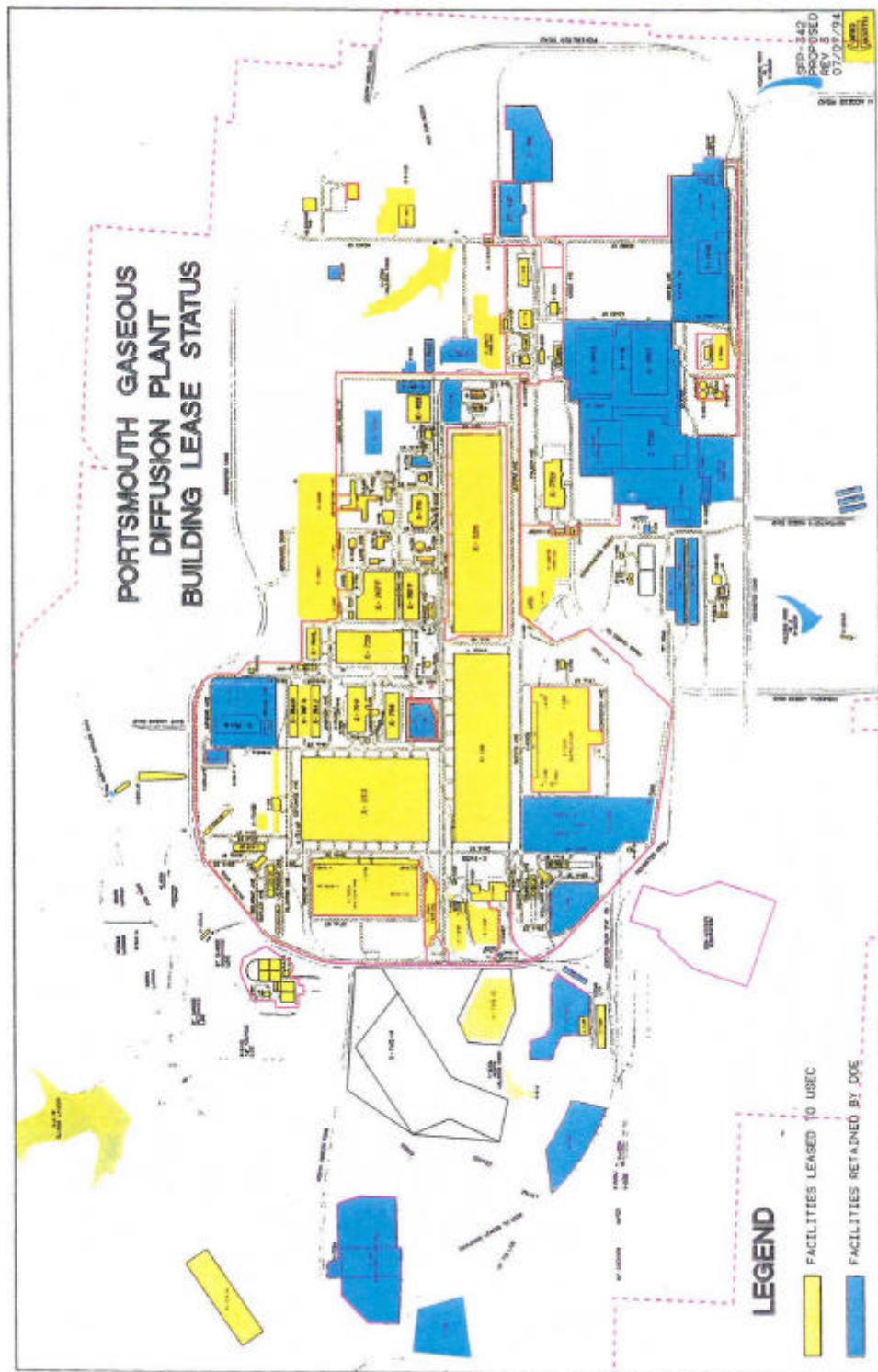
**Regional Location of PORTS**



**LEGEND**

★ PORTSMOUTH GASEOUS DIFFUSION PLANT

Figure 2.1-2



The gaseous diffusion enrichment process takes place in three large process buildings; X-333, X-330, and X-326. Process buildings X-333 and X-330 were built in 1955 and used for the initial and intermediate phases, respectively, of uranium enrichment. The process building (X-326), which was built in 1956, was originally used for the high enrichment phase, but is no longer used for this purpose. The X-326 is currently used for product withdrawal and side feeding. In addition, from early 1997 to mid 1998 the X-326 product withdrawal equipment was used for HEU blending activities.

Various  $\text{UF}_6$  feed, withdrawal, and sampling systems and  $\text{UF}_6$  cylinder operations are located in the three process buildings, as well as the X-342A Feed Vaporization and Fluorine Generation Facility, X-343 Feed Vaporization and Sampling Facility, and the X-344A Toll Enrichment Services Facility.

Three large facilities, the X-700 Converter Shop and Cleaning Building, X-705 Decontamination and Recovery Facility, and the X-720 Maintenance and Stores Building provide most of the equipment maintenance support for the diffusion cascade. Equipment removed from the cascade is disassembled and decontaminated in X-705, which also houses equipment/systems for the recovery of uranium from decontamination solutions.

## **2.2 Key Uranium Processing Facilities**

Four major facilities were suspected of containing processes where RU constituents may be concentrated and, thereby, present the potential for worker exposure or environmental contamination. These facilities are listed below and their description and processes are described in subsequent sections:

1. X-344 Feed Manufacturing Plant;
2. Cascade (X-333, X-330, and X-326) and associated feed, withdrawal and sampling facilities;
3. X-705 Decontamination and Recovery Facility; and
4. X-705 Oxide Conversion Facility.

### **2.2.1 Feed Manufacturing Plant**



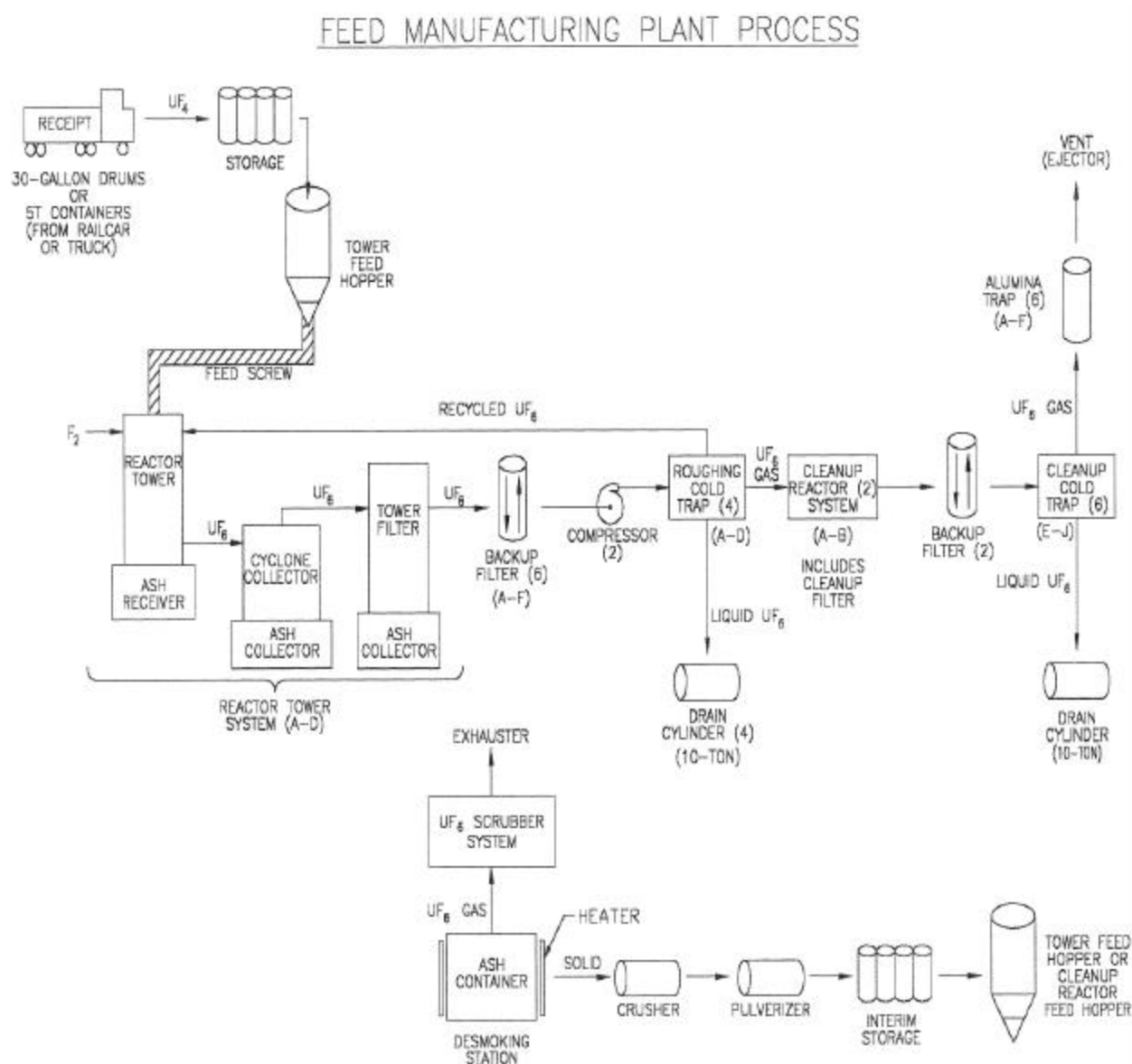
X-344 FEED MANUFACTURING FACILITY

### 2.2.1.1 Plant Description

The X-344 Feed Manufacturing Plant was located largely in what is now the X-342A Feed Vaporization and Fluorine Generation Building and X-344A Toll Enrichment Services Facilities, which are located north of the X-745B Toll Enrichment Process Gas Yard in the north-central region of the PORTS site. The feed plant was constructed and operated in the early days for the production of  $UF_6$  from  $UF_4$  green salt. The plant consisted of: 1) a building to house the process equipment, offices, and maintenance areas; 2) an auxiliary building for ash storage and acid neutralization; 3) an acid unloading and storage facility; and 4) modifications to the existing X-342A.

The process for producing  $UF_6$  was by direct fluorination of  $UF_4$  in a tower reactor as shown in Figure 2.2.1.1-1. The product  $UF_6$  was piped to refrigerated cold traps where it was separated from other gases by condensation. Subsequently, the cold traps were isolated, heated, and the  $UF_6$  drained as a liquid into 10-ton cylinders for eventual vaporization into enrichment cascades.

Figure 2.2.1.1-1



The large quantities of fluorine required for the process were produced by electrolysis of hydrogen fluoride (HF) in fluorine cells. The HF was stored in three 12,500 gallon storage tanks located within a concrete diked area protected by a Butler-type HF storage building (X-344C) located next to a spur track northeast of the X-344 building. Pumps were provided to transfer HF from the storage tanks to vaporizers located in the X-344 and then to the fluorine cells. A total of 40 cells were available for fluorine production. Equipment was provided to filter the fluorine and remove any residual electrolyte, and heat it prior to being metered into the tower reactor system(s).

Handling equipment was provided for receiving green salt powder in 30-gallon drums or five-ton containers in railroad cars or trucks. Facilities were provided for washing and drying of empty 30-gallon drums. A means was provided for adding one 30-gallon drum of ash to each five-ton container prior to its rotation and positioning over the tower feed hoppers.

The green salt fluorination system consisted of four tower reactors and the fluorine cleanup reactors. Each reactor system included a fluorine preheater, feed hopper, feed screw, reactor tower, cyclone barrier filter, and ash receiver similar in design to that used in the Oak Ridge feed plant. Following the reactor tower systems, the  $UF_6$  gas was collected and piped to a backup filter station prior to being compressed and passed through one of four roughing cold traps. A portion of the gas from the cold traps was recycled back to the reactor towers and a portion was piped to a cleanup reactor system to remove any residual fluorine. The gas from the cleanup reactors was passed through one of the six cleanup cold traps. Any uncondensed  $UF_6$  in the gas stream from the cleanup cold traps was removed in the alumina traps prior to venting to the environment.

Four drain positions were equipped for collecting the liquid  $UF_6$  drained from the cold traps. Each position was designed to handle a 10-ton cylinder.

Ash grinding and storage facilities were provided for storing and processing unreacted green salt collected in the ash receivers under the towers, filters, and cyclones. These facilities were equipped for heating, crushing, pulverizing, and containerizing the ash for blending with fresh green salt for use in the tower reactors.

A system similar to that used in the Paducah feed plant was provided for neutralizing waste acid from the water scrubbing system before the water was passed to the sewer. This system provided for the mixing of lime with the acid solution.

Maintenance areas were provided for dismantling and repairing fluorine cells and other process equipment.

#### **2.2.1.2 Material Flowsheet**

The materials flowsheet for feed manufacturing is shown in Figure 2.2.1.2-1.

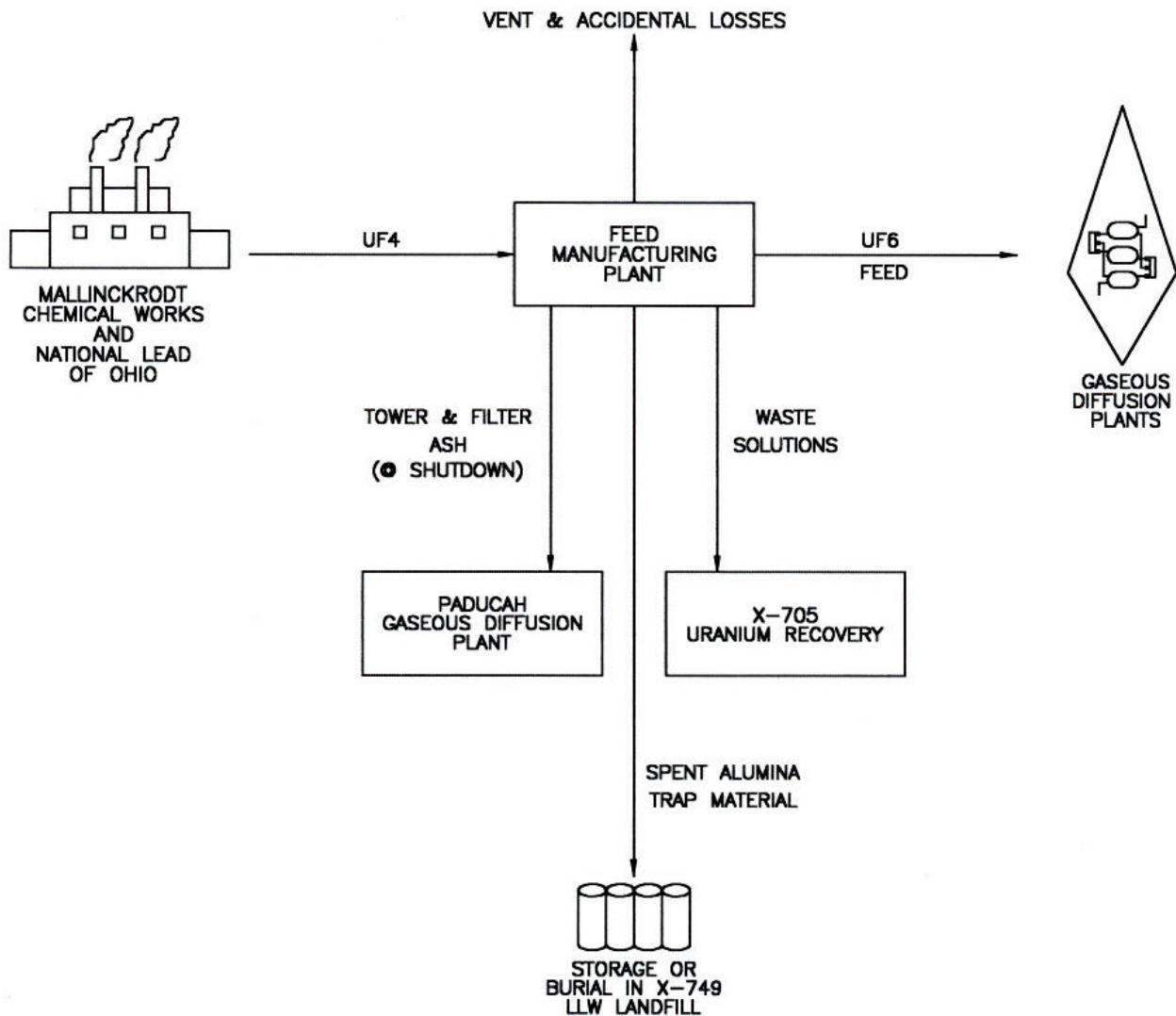
A review of plant records (Ref. 3 and 4) indicates that all  $UF_4$  fed into the feed manufacturing plant during its history was of normal enrichment from two sources; namely Mallinckrodt Chemical Works (MCW) and National Lead of Ohio (NLO). No record can be found of  $UF_4$  produced from RU forms ever having been fed in the feed manufacturing plant. The  $UF_6$  product was stored and either fed to the PORTS enrichment cascade or shipped to one of the other enrichment cascades. The reaction of  $UF_4$  and fluorine in the tower reactors did not consume all of the  $UF_4$  and some of it was caught in the ash receivers under the towers, filters and cyclones. The ash, which contained useable uranium, was recycled through the tower after further processing. The ash receivers were stored for approximately two months to allow the radiation level to drop to the point where the ash could be handled. Following the storage period, the ash was heated and the absorbed  $UF_6$  driven off and recovered in a water scrubber system. The  $UF_6$  free ash was then crushed, screened, pulverized and containerized for future blending with fresh green salt for use in the tower reactors. The only time ash was removed from the plant was after shutdown when it was pulverized, containerized and shipped to Paducah.

The waste acid from the water scrubbing system was neutralized with lime and permitted to pass into the sewer or containerized and transferred to the X-705 for processing.



Figure 2.2.1.2-1

## FEED MANUFACTURING PLANT MATERIAL FLOWSHEET



The spent alumina from the chemical traps was containerized and either stored in the X-744G Bulk Storage Facility or buried in the X-749 Low Level Radioactive Waste Burial Ground.

### 2.2.1.3 Operating History

The Feed Manufacturing Plant was turned over to Goodyear Atomic Corporation (GAT) on April 25, 1958. Initial testing of the plant was performed using a special allotment of  $UF_4$  shipped from Mallinckrodt Chemical Works for the production of  $UF_6$  standards. The initial testing was completed on May 14, 1958 and the plant started production operations on May 15, 1958. The plant continued operations until February 1962 at which time it was shut down.

During its 46-1/2 month operational life 11,983 MTU of  $UF_4$  was fed, at an average rate of 9.5 MTU per day, producing a total of 11,890 MTU of  $UF_6$  (see Appendix I). This operation experienced numerous operating and maintenance problems resulting in significant radionuclide emissions to the atmosphere as well as contributing to its shutdown. The plant lost an average of 407 kgU per year to the atmosphere from 1959 until its shutdown in 1962, (Ref. 5).

At shutdown all material inventory was removed from the system, containerized and moved to storage for final disposition. About 23.3 MTU as ash was removed, pulverized containerized and shipped to Paducah for processing.

### 2.2.1.4 Current Status

After shutdown in 1962 and material removal, the process equipment was dismantled. In the early 1970's, new plans for the X-344 facility were prepared and the building was converted by 1975 to serve as the shipping/receiving point for low-assay  $UF_6$  (less than 5%). Currently the facility is used for sampling 10-ton product cylinders and transferring product into smaller (2-1/2 ton) customer-owned cylinders for shipment. Use of all but four fluorine generation cells was discontinued with the remaining fluorine cells used for maintenance spares.

## 2.2.2 Cascade and Feed Facilities



South Side of X-333 Process Building



### 2.2.2.1 Description

The PORTS cascade is comprised of 4080 stages of process equipment and is capable of enriching uranium to 97%  $^{235}\text{U}$  assay. Table 2.2.2.1-1 summarizes the stage arrangements per building and itemizes the number of stages of the various size equipment. The 33-size equipment located in the X-333 Process Building is the largest. Figure 2.2.2.1-1 shows a typical cell flow diagram for X-33-size equipment. The 25-size equipment located in the X-326 Process Building is the smallest. The cascade originally was fed from the X-342 building which housed 12 steam vaporizer bays used to heat 2-1/2 and 10-ton  $\text{UF}_6$  cylinders. Two product withdrawal facilities located in the X-326 building, one being for Very Highly Enriched (VHE) product and one for Extended Range Product (ERP) for lower assays and one tails (depleted stream) withdrawal facility located in the X-330 building were part of the original plant design. A purge facility used to vent light gases is also located in the top (i.e., near VHE product areas) cascade of the X-326 building.

**Table 2.2.2.1-1**

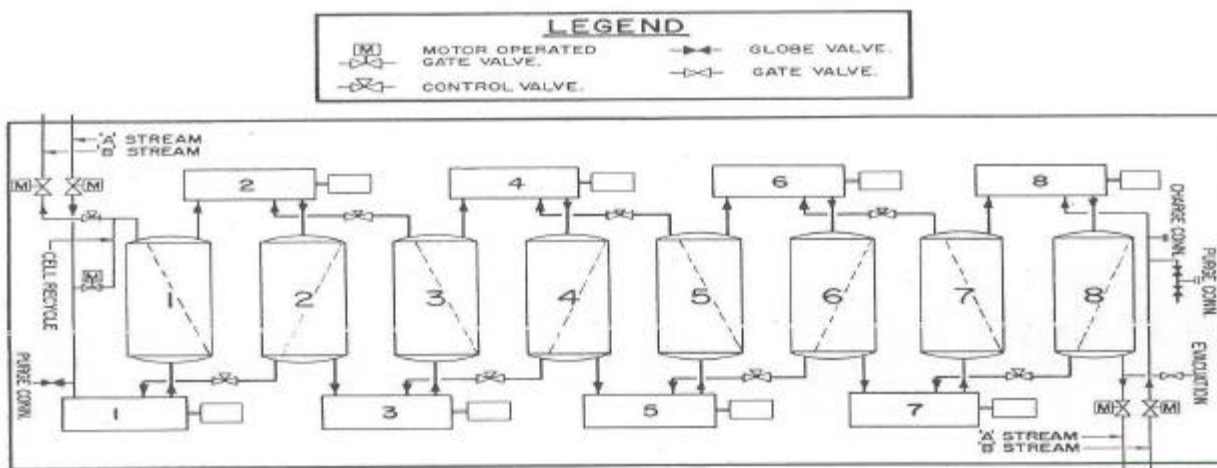
**Cascade Configurations**

| Process Building | Equipment Size | Number of Stages |
|------------------|----------------|------------------|
| X-333            | 33 or (000)    | 640              |
| X-330            | 31 or (00)     | 500              |
|                  | 29 or (0)      | 600              |
| X-326            | 27             | 720              |
|                  | 25             | 1620             |
| <b>TOTAL</b>     |                | <b>4080</b>      |

Through the years, PORTS' mission evolved from high assay production for military uses to providing low enrichment services for fuel to be used in commercial nuclear power plants. Additional facilities have been built and existing facilities have been modified to reliably support the new mission. A new feed facility, the Feed

**Figure 2.2.2.1-1**

**Cell Flow Diagram for X-33 Units**



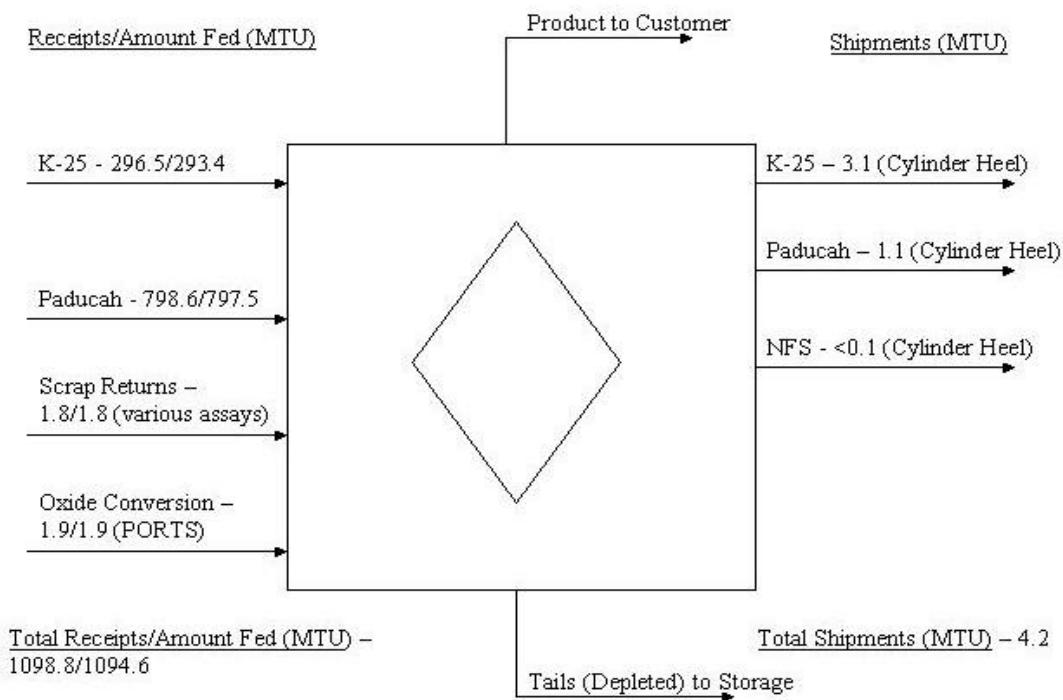
Vaporization and Sampling Building (X-343), was built and placed on line in 1983 and houses seven autoclaves. The 12 steam chests in X-342 were removed and two new autoclaves were installed to feed the cascade. An additional Low Assay Withdrawal (LAW) facility was installed in the X-333 building and modifications were made to enable product withdrawal at the tails withdrawal area, as well as tails withdrawal at LAW. The X-344 facility was modified into a toll enriching facility where product transfers from 10-ton processing cylinders to 2-1/2 ton customer cylinders with associated sampling is accomplished and cylinders are loaded onto trucks/railcars and shipped off-site.

#### 2.2.2.2 Material Flowsheet

See Material Flowsheet – Cascade – RU-UF<sub>6</sub>, Figure 2.2.2.2-1

**Figure 2.2.2.2-1**

#### **Material Flowsheet – Cascade RU-UF<sub>6</sub>**



#### 2.2.2.3 Feed Specifications

1. Feed <5% - American Society for Testing Materials (ASTM) specification C-787-96 (or previous revisions) for natural uranium and UF<sub>6</sub> that has been received from irradiated uranium which has been reprocessed and converted to UF<sub>6</sub> (see Ref. 6 and 7).
  - Primary specification item – total alpha activity from Np and Pu to be limited to 1,500 disintegrations per minute/gram uranium (dpm/gU).

2. Feed >5% assay – scrap return program beginning in 1968 that had reactor return constituents was under a 1961 specification stating that total activity level to be less than 1,500 dpm/gU. Although these specifications were reissued in April 1971 by the AEC which raised the alpha activity level to 15,000 dpm/gU, PORTS continued to use 1,500 dpm/gU limit for scrap acceptance except for approved deviations on the NLO material received at 3,000 dpm/gU.
3. Shipments between the diffusion plants are exempt from ASTM specification criteria, although no known receipts or shipments have been identified that exceed the total of 1,500 dpm/gU limit.

#### 2.2.2.4 Product Specifications

1. Product <5% - DOE memo dated January 22, 1993 and ASTM specification C-996-96 (or previous revision) (see Ref. 7 and 8).
2. Product ≥5% - military uses exempt from ASTM specifications. Naval and weapons program specifications were used.

#### 2.2.2.5 Operating History

Startup of the PORTS cascade began early in FY 1955. Appendices II and III summarize the total uranium and RU received and shipped from PORTS (all forms) from startup to March 31, 1999. Appendix IV shows all the UF<sub>6</sub> fed to the cascade and its source, including reactor returns on an annual basis over the same 44-year timeframe. Reactor returns were fed to the cascade in timeframes shown in Table 2.2.2.5-1.

**Table 2.2.2.5-1**

#### **REACTOR RETURN CASCADE FEEDS (UF<sub>6</sub>)**

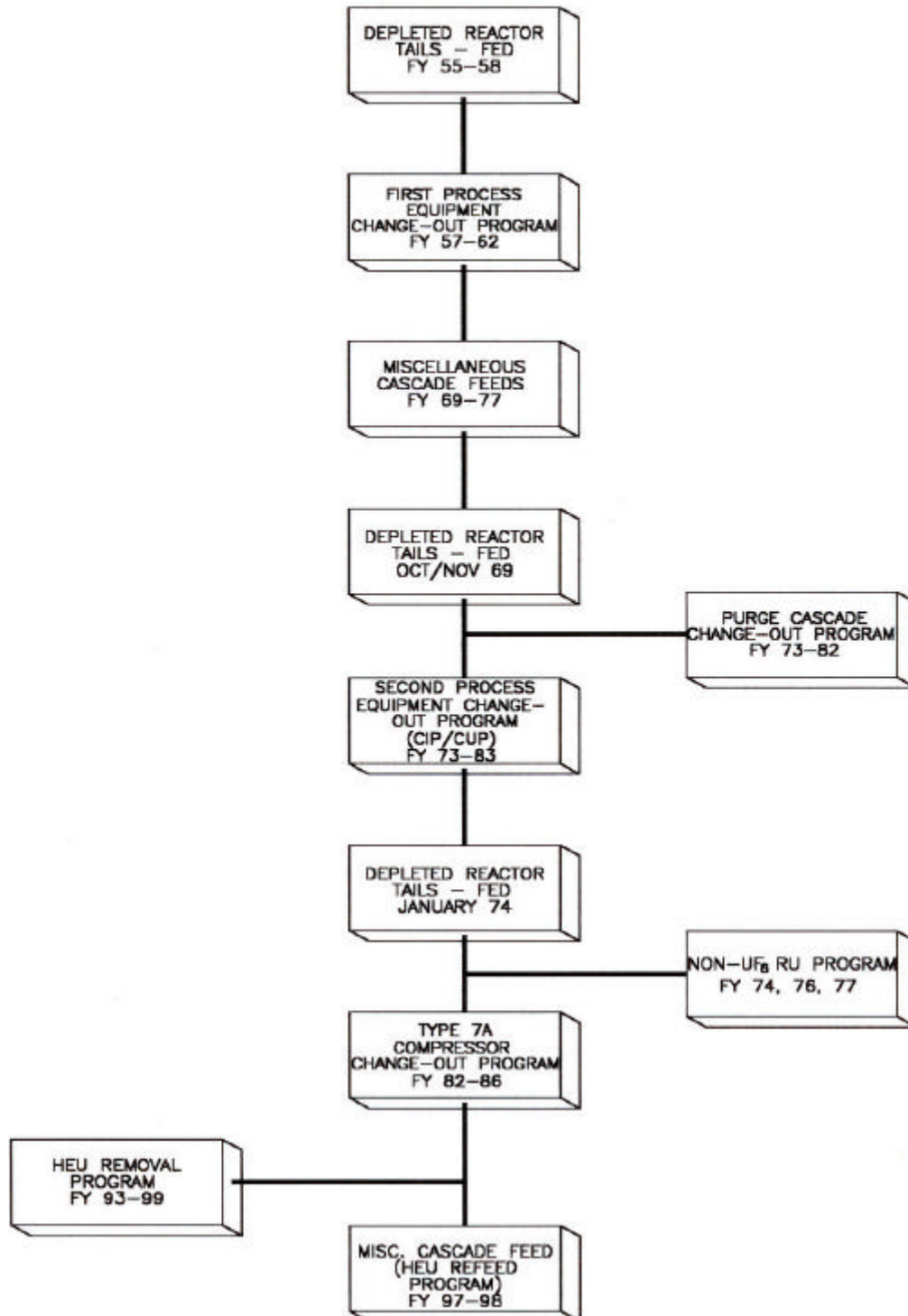
| Fiscal Year | Amount Fed MTU | Assay %U <sup>235</sup> | Source  | Remarks              |
|-------------|----------------|-------------------------|---|----------------------|
| 1955        | 105.8          | 0.64-0.68               | Paducah   | Fed May – Sept. 1955 |
| 1956        | 54.5           | 0.64-0.68               | Paducah   |                      |
|             | 293.4          | 0.64-0.68               | Oak Ridge   |                      |
| 1957        | 6.2            | 0.64-0.68               | Paducah   |                      |
| 1958        | 64.2           | 0.64-0.68               | Paducah   |                      |
| 1970        | 168.1          | 0.64-0.68               | Paducah   | Fed Oct. & Nov. 1969 |
| 1974        | 398.8          | 0.64-0.68               | Paducah   | Fed Jan. 1974        |
| 1968-1977*  | 0.15           | 78-80                   | Division of International Affairs                 |                      |
| 1977-1998*  | 0.15           | 78-97                   | Babcock & Wilcox                                  |                      |
| 1969-1993*  | 0.07           | 78                      | USAEC Office of Safeguards & Materials Management |                      |
| 1997-1998   | 1.10           | 56-82                   | France  |                      |
|             | 0.33           | 80                      | NUMEC   |                      |
| 1974-1978*  | 1.86           | 2-50                    | PORTS Oxide Conversion                            |                      |

\*Streams will require additional research to pinpoint the feed date if deemed necessary.

The operating history will be discussed with focus on significant RU events shown in Figure 2.2.2.5-1. These same events will be portrayed as campaigns in Section 5.

Figure 2.2.2.5-1

SIGNIFICANT RECYCLED URANIUM EVENTS  
— CHRONOLOGY —



During plant startup, some RU that had been converted to  $UF_6$  at Paducah and Oak Ridge was fed to the cascade. Five hundred twenty-seven (527) MTU RU of a total feed of 33,356 MTU (a total of about 1.6%) was fed during the FY 1955 - FY 1958 timeframe. Feed cylinder numbers, assay, date fed, and cascade feed points have been located, analyzed and tabulated (see Appendix V through VII). The Oak Ridge feed (296.5 MTU) came in 2-1/2 ton cylinders, whereas the Paducah material (230.6 MTU) came in both 2-1/2 ton and 10-ton cylinders. It appears that some of the 200 2-1/2 ton cylinders used were cycled between the plants with RU materials two to four times, thereby concentrating the RU in the cylinder heels.



Maintenance Workers Finishing Assembly of a X33 Size Converter

Following this period of RU feeds, the plants' first equipment change-out program commenced. This converter barrier replacement program ran from FY 1957-FY 1962 during which time 560 stages of X-33-size, 500 stages of X-31 size, and 280 stages of X-29 size converter barrier were replaced. The amount of TRU in the RU that could have been removed from the cascade and the process equipment during decontamination in this timeframe is based upon the concentration data provided in Refs. 2 and 2a, and for this period is calculated as 32 g Np and 2.1 mg Pu.

Reactor returns were again received from Paducah in FY 1968 – FY 1969 (568 MTU). One hundred sixty-eight (168) MTU was fed to the cascade in October and November 1969 (FY 1970), and 400 MTU (which is the largest amount fed at PORTS in any one year, as well as comprising 35% of all RU fed to the plant to date) was fed in January 1974. A manual  $UF_6$  product cylinder history card system giving cylinder transactions since startup is available in the USEC Nuclear Materials Accounting Department for cylinders located at PORTS. Some other interplant cylinder history is also available. From these records, it appears that the 26 10-ton cylinders fed in FY 1970 were received, fed, and filled with PORTS tails, and returned. The 62 10-ton cylinders fed in FY 1974 were returned to Paducah empty where 28 were cleaned and returned to PORTS empty in the June 1975 timeframe.

Following this period of RU feeds, the plant's second major equipment change-out cascade improvement program (CIP) began in FY 1973 and was completed in FY 1983. Essentially all X-33 size (640 stages) and X-31 size (500 stages) of process equipment and piping were removed, decontaminated, and modified. The amount of

TRU that could have been removed from the cascade during this campaign is based on the concentration data provided in Refs. 2 and 2a, and for the period is calculated as 12.8 g Np and 1.3 mg Pu.

During the period of FY 1968 - FY 1978, small quantities of various assays RU were received as UF<sub>6</sub> or converted to UF<sub>6</sub> at PORTS' Oxide Conversion Facility. This was part of the government's scrap returns program. See scrap returns program (UF<sub>6</sub>) and converted oxides for cascade feed, Table 2.2.2.5-2, for amount of RU-UF<sub>6</sub> received per shipper, amount of RU-UF<sub>6</sub> produced at PORTS oxide conversion, assay, amount fed through March 1999, and the amount in storage as of March 31, 1999. Detailed information is available for this UF<sub>6</sub>, except for the specific dates cylinders were fed. A total of 1.4 MTU was fed to the cascade in the X-326 Building during the HEU refeed program in the FY 1997 - FY 1998 timeframe was from France and NUMEC; UF<sub>6</sub> assays ranged from 56-82% (see Appendices VIII through XII).

**Table 2.2.2.5-2**  
**SCRAP RETURN PROGRAM (UF<sub>6</sub>) AND CONVERTED OXIDES**  
**FOR CASCADE FEED**

| Shipper  |  | Assay % | Fiscal Year Received/Converted | Amount of UF <sub>6</sub> Received/Converted (MTU) | No. & Sizes of Cylinders | Timeframe Fed to Cascade (FY) | Amount Fed MTU) | In Storage as of March 31, 1999 (MTU) | Rejected & Returned (MTU) |
|--|--|---------|--------------------------------|--|--------------------------|-------------------------------|-----------------|---------------------------------------|---------------------------|
| Division of International Affairs                  |  | 78-80   | 1968                           | 0.15   | 10-5"                    | 1968-77                       | 0.15            | 0                                     | --                        |
| Babcock & Wilcox                                   |  | 78-97   | 1977                           | 0.15   | 11-5"                    | 1977-78                       | 0.15            | 0                                     | --                        |
| USAEC Office of Safeguards and Material Management |  | 1.35    | 1969                           | 2.73   | 2<br>(2-1/2 ton)         | 1969-93                       | --              | 2.73                                  | --                        |
|  |  | 78      |                                | 0.07   | 3 (5")                   |                               | 0.07            | 0                                     | --                        |
| France   |  | 56-82   | 1972-78                        | 1.6  | 67-5"                    | 1997-98                       | 1.1             | 0                                     | 0.3                       |
| NUMEC  |  | 80      | 1972                           | 0.33   | 20-5"                    | 1997-98                       | 0.33            | 0                                     | --                        |
| Oxide Conversion at PORTS from:                    | Idaho Chemical Processing Plant (ICPP) | 50      | 1974 (Jan. & Feb.)             | 1.4  | Unknown                  | 1974-77                       | 1.4             | 0                                     | --                        |
|  | NLO                                    | 2.9     | 1976 (Jan. & May)              | 4.2  | 13<br>(5" & 12")         | 1976-77<br>(10 cyl.)          | 0.46            | 0.12<br>(3 cyl.)                      | --                        |
|  |  |         |                                |  | 4 (2-1/2 ton)            | 0                             | --              | 3.62                                  | --                        |

Throughout the plant's history, many pieces of process equipment have been changed out under routine plant maintenance in addition to the two major upgrade programs discussed above (see Annual Equipment Replacement Summary Table 2.2.2.5-3). Two smaller equipment change-outs were also reviewed and are discussed below.

The purge cascade converters began to plug with <sup>99</sup>Tc compounds and other metallic impurities from feeds. A program to remove, dismantle, and clean approximately 25 converters occurred during FY 1973-FY 1978. An estimated 0.375 kg of <sup>99</sup>Tc was removed from the cascade with this program. An off-stream hot-gas treatment



process was developed that allowed some  $^{99}\text{Tc}$  plugged converters to be unplugged in place. The same process was used to clean equipment of  $^{99}\text{Tc}$  prior to maintenance/equipment removal activities.

**Table 2.2.2.5-3**

**ANNUAL EQUIPMENT REPLACEMENT SUMMARY**

| Calendar Year | Compressor Removals |      |      |      |      |       | Converter Removals |      |      |      |      | Total |
|---------------|---------------------|------|------|------|------|-------|--------------------|------|------|------|------|-------|
|               | X-33                | X-31 | X-29 | X-27 | X-25 | Total | X-33               | X-31 | X-29 | X-27 | X-25 |       |
| 1954          | --                  | 47   | --   | --   | --   | 47    | --                 | 6    | --   | --   | --   | 6     |
| 1955          | 2                   | 9    | 7    | 14   | 8    | 40    | 1                  | 1    | 4    | 2    | 12   | 20    |
| 1956          | 20                  | 10   | 7    | 13   | 13   | 63    | 12                 | 59   | 2    | 0    | 2    | 75    |
| 1957          | 30                  | 18   | 2    | 17   | 7    | 74    | 169                | 304  | 0    | 0    | 1    | 474   |
| 1958          | 22                  | 80   | 18   | 16   | 8    | 144   | 345                | 15   | 1    | 2    | 0    | 363   |
| 1959          | 28                  | 106  | 2    | 8    | 8    | 152   | 208                | 0    | 2    | 0    | 0    | 210   |
| 1960          | 8                   | 203  | 54   | 12   | 9    | 286   | 40                 | 20   | 213  | 2    | 0    | 275   |
| 1961          | 6                   | 80   | 257  | 17   | 10   | 370   | 63                 | 40   | 40   | 0    | 0    | 143   |
| 1962          | 0                   | 102  | 70   | 4    | 12   | 188   | 5                  | 50   | 0    | 0    | 0    | 55    |
| 1963          | 12                  | 50   | 38   | 5    | 6    | 111   | 17                 | 20   | 20   | 0    | 0    | 57    |
| 1964          | 5                   | 42   | 45   | 5    | 12   | 109   | 4                  | 20   | 30   | 1    | 0    | 55    |
| 1965          | 8                   | 23   | 95   | 10   | 24   | 160   | 5                  | 20   | 22   | 0    | 0    | 47    |
| 1966          | 4                   | 37   | 15   | 13   | 14   | 83    | 5                  | 10   | 0    | 12   | 2    | 29    |
| 1967          | 19                  | 4    | 2    | 10   | 15   | 50    | 26                 | 0    | 0    | 0    | 0    | 26    |
| 1968          | 6                   | 1    | 2    | 12   | 18   | 39    | 20                 | 0    | 0    | 0    | 0    | 20    |
| 1969          | 22                  | 1    | 26   | 3    | 20   | 72    | 20                 | 1    | 0    | 0    | 0    | 21    |
| 1970          | 21                  | 1    | 20   | 2    | 6    | 50    | 1                  | 14   | 0    | 0    | 0    | 15    |
| 1971          | 15                  | 1    | 12   | 2    | 5    | 35    | 12                 | 1    | 0    | 0    | 1    | 14    |
| 1972          | 36                  | 10   | 18   | 4    | 6    | 74    | 12                 | 0    | 0    | 0    | 8    | 20    |
| 1973          | 24                  | 0    | 10   | 5    | 10   | 49    | 30                 | 0    | 0    | 0    | 6    | 36    |
| 1974          | 40                  | 4    | 13   | 3    | 5    | 65    | 44                 | 0    | 0    | 0    | 0    | 44    |
| 1975          | 127                 | 42   | 0    | 6    | 10   | 185   | 119                | 0    | 0    | 0    | 1    | 120   |
| 1976          | 148                 | 11   | 16   | 5    | 8    | 188   | 118                | 1    | 1    | 0    | 20   | 140   |
| 1977          | 198                 | 66   | 5    | 2    | 12   | 283   | 150                | 40   | 0    | 0    | 0    | 190   |
| 1978          | 150                 | 70   | 2    | 5    | 10   | 237   | 148                | 52   | 0    | 0    | 4    | 204   |
| 1979          | 54                  | 40   | 8    | 6    | 18   | 353   | 54                 | 30   | 0    | 0    | 0    | 84    |
| 1980          | 106                 | 3    | 4    | 15   | 15   | 143   | 110                | 6    | 0    | 0    | 18   | 134   |
| 1981          | 12                  | 197  | 12   | 7    | 21   | 249   | 6                  | 154  | 84   | 0    | 0    | 244   |
| 1982          | 4                   | 190  | 5    | 34   | 23   | 256   | 2                  | 190  | 16   | 1    | 18   | 227   |
| 1983          | 25                  | 40   | 6    | 42   | 25   | 138   | 4                  | 8    | 0    | 0    | 0    | 12    |
| 1984          | 32                  | 6    | 6    | 54   | 13   | 111   | 14                 | 0    | 0    | 0    | 0    | 14    |
| 1985          | 44                  | 10   | 2    | 38   | 20   | 114   | 14                 | 2    | 0    | 0    | 0    | 16    |
| 1986          | 21                  | 2    | 0    | 18   | 3    | 44    | 20                 | 2    | 0    | 0    | 0    | 22    |
| 1987          | 2                   | 2    | 2    | 11   | 23   | 40    | 18                 | 0    | 0    | 0    | 1    | 19    |
| 1988          | 2                   | 2    | 3    | 7    | 23   | 37    | 18                 | 1    | 0    | 0    | 0    | 19    |

MgF<sub>2</sub> sidestream traps to remove  $^{99}\text{Tc}$  were installed for use with cells X25-7-15, 17, and 19. These cells correspond to peak concentrations of  $^{99}\text{Tc}$  in the cascade. This trapping system was successful in reducing the cascade  $^{99}\text{Tc}$  inventory, but created a source of concentrated  $^{99}\text{Tc}$  trap media. The quantity of  $^{99}\text{Tc}$  in this media has not been quantified.

Another program consisted of the removal and subsequent decontamination and rebuilding of approximately 190 Fairchild type 7A compressors during FY 1982-FY 1986. These compressors were located in the X-326 building and are part of the X-27 size equipment. Due to a design problem, these compressors had wet air leakage at the bolted compressor flange area. No TRU was considered to be removed with this activity.

In FY 1991, high assay production was terminated and 1,680 stages of equipment were shut down. Treatment for deposit removal, as needed, and mothballing of these shutdown cells occurred between FY 1993 and FY 1998. A total of 240 X-27 size isotopic stages and 180 X-25 size purge cascade stages remain in operation

In July 1993, USEC leased the enrichment facilities from DOE with Lockheed Martin Utility Services becoming the Maintenance and Operating (M&O) contractor.

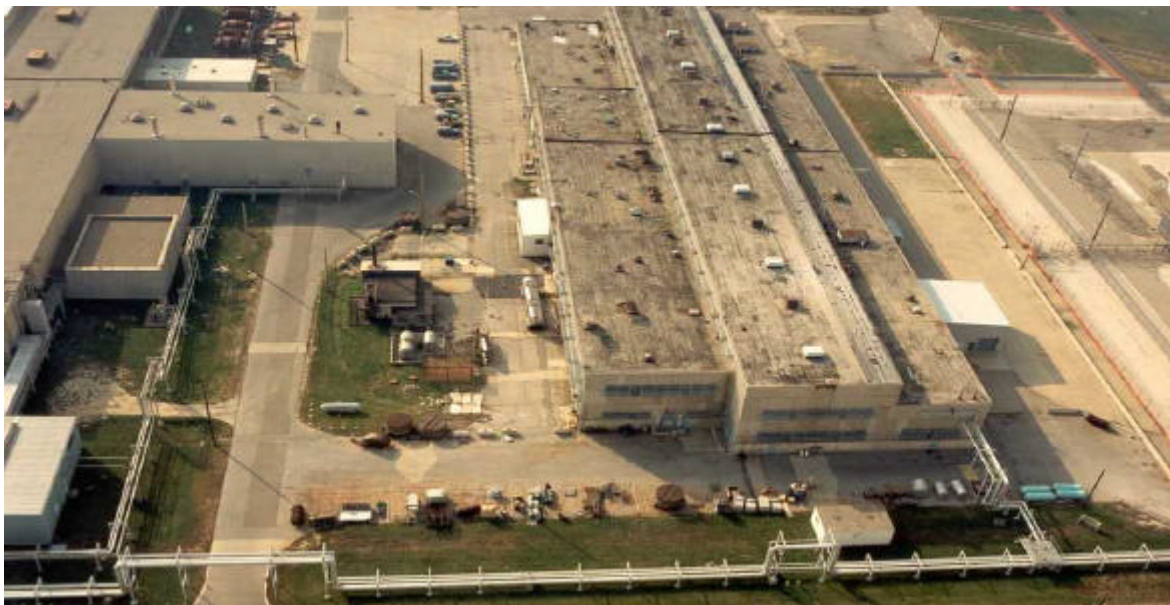
During FY 1997 - FY 1998, HEU  $UF_6$  stored on site was fed and blended to LEU specifications in an agreement between DOE and USEC as part of a program to reduce PORTS inventory of HEU.

In March 1997, regulatory oversight of enrichment operations transferred from DOE to the Nuclear Regulatory Commission (NRC). In December 1998, X-326 side purge cell X25-7-2 caught fire due to internal compressor rubbing and resultant exothermic reaction. Three other side purge cells were damaged and are undergoing rework/rebuild. In May 1999, USEC took over direct operation of enrichment facilities.

#### **2.2.2.6 Current Status**

USEC is currently leasing and operating the cascade and its support facilities. The X-326 is essentially shut down except for 240 X-27 size isotopic stages and 180 X-25 size purge cascade stages. Varying amounts of X-27 size equipment and X-25 size equipment are running. Efforts are under way to rebuild the side purge cascade cells damaged from a 1998 fire. The shutdown equipment has been cleaned of large deposits and mothballed. All X-333 and X-330 building equipment is available for USEC's use. Approximately 88% of the X-33 size equipment, 60% of X-31 size equipment, and 8% of X-29 size equipment are running as of April 2000. The HEU- $UF_6$  in storage has been refed/down-blended and other uranium bearing materials of greater than 20% assay have been shipped off site. Six cylinders with RU- $UF_6$  (6.5 MTU) were in storage as of March 31, 1999 at PORTS.

#### **2.2.3 Uranium Recovery**



X-705 Decontamination and Uranium Recovery Facility

### 2.2.3.1 Plant Description

The uranium recovery operation is contained primarily in the X-705 with ancillary waste treatment operations located in the X-700 and at various times at the X-701B Holding Pond. A pictorial diagram of the integrated uranium recovery process is shown in Figure 2.2.3.1-1. The oxide conversion segment of this process will be discussed as a separate plant in Section 2.2.4 due to its unique mission.

The uranium recovery facility was designed to reclaim uranium from the following principle sources:

1. Decontamination solutions from equipment removed from the cascade and areas (small and large parts)
2. Field decontamination solutions
3. Trap media leaching
4. Oxide conversion,  $\text{UF}_4$  conversion, and incinerator ashes and filtrates
5.  $\text{UF}_6$  cylinder cleaning
6. Laboratory and miscellaneous sources.

The processes used to create recoverable quantities of uranium from the above sources are varied in size, complexity, location, and throughput. They all, however, involve chemical or mechanical removal of uranium compounds from metallic surfaces and the subsequent dissolving of the removed uranium in aqueous or acidic solutions. These operations do not themselves selectively concentrate TRU or FP, but rather maintain the relative input concentrations of the constituents of concern. Concentration of the constituents in total is realized as solvents are reused/recycled until saturated.

The process used to recover uranium from solutions has been referred to as solution recovery, counter current extraction, or solvent extraction and appears to be a variant of the Plutonium and Uranium Extraction (PUREX) process developed for use at Hanford.

In this process (Figure 2.2.3.1-2), uranyl solutions are first evaporated to increase the specific gravity. The solution is then fed, along with nitric acid, to an extraction column where the uranyl ion is selectively captured by a mixture of tributyl phosphate (organic solvent) and Varsol (petroleum distillate). Next, this mixture has the uranyl ion as a nitrate stripped from it in a second (stripping) column with deionized water. The solvent solution is recycled to the extraction column for reuse. The aqueous uranyl nitrate solution (product) is fed to an evaporator where excess water is evaporated. The dewatered solution is then fed to a calciner (rotary kiln) where it is denitrated and further dried to produce  $\text{U}_3\text{O}_8$ , the final product, for storage, shipment, or fluorination to  $\text{UF}_6$ . Waste from this uranium recovery process is principally the depleted acid as raffinate from the extraction column.

TRUs in the uranyl solutions appear to follow the uranyl ions throughout all steps of the oxide formation process. The minor amounts that accompany the raffinate are discussed later. Experimental data collected and published by Walker (circa 1977) (Ref. 9) are as follows:

| Process Step              | Total TRU | $^{237}\text{Np}$ | $^{239-240}\text{Pu}$ | $^{238}\text{Pu}$ |
|---------------------------|-----------|-------------------|-----------------------|-------------------|
| Loaded solvent (dpm/ml)   | 60,700    | 16,900            | 23,700                | 20,100            |
| Stripped solvent (dpm/ml) | 675       | 222               | 230                   | 223               |
| % stripped                | 98.89     | 98.69             | 99.03                 | 89.98             |

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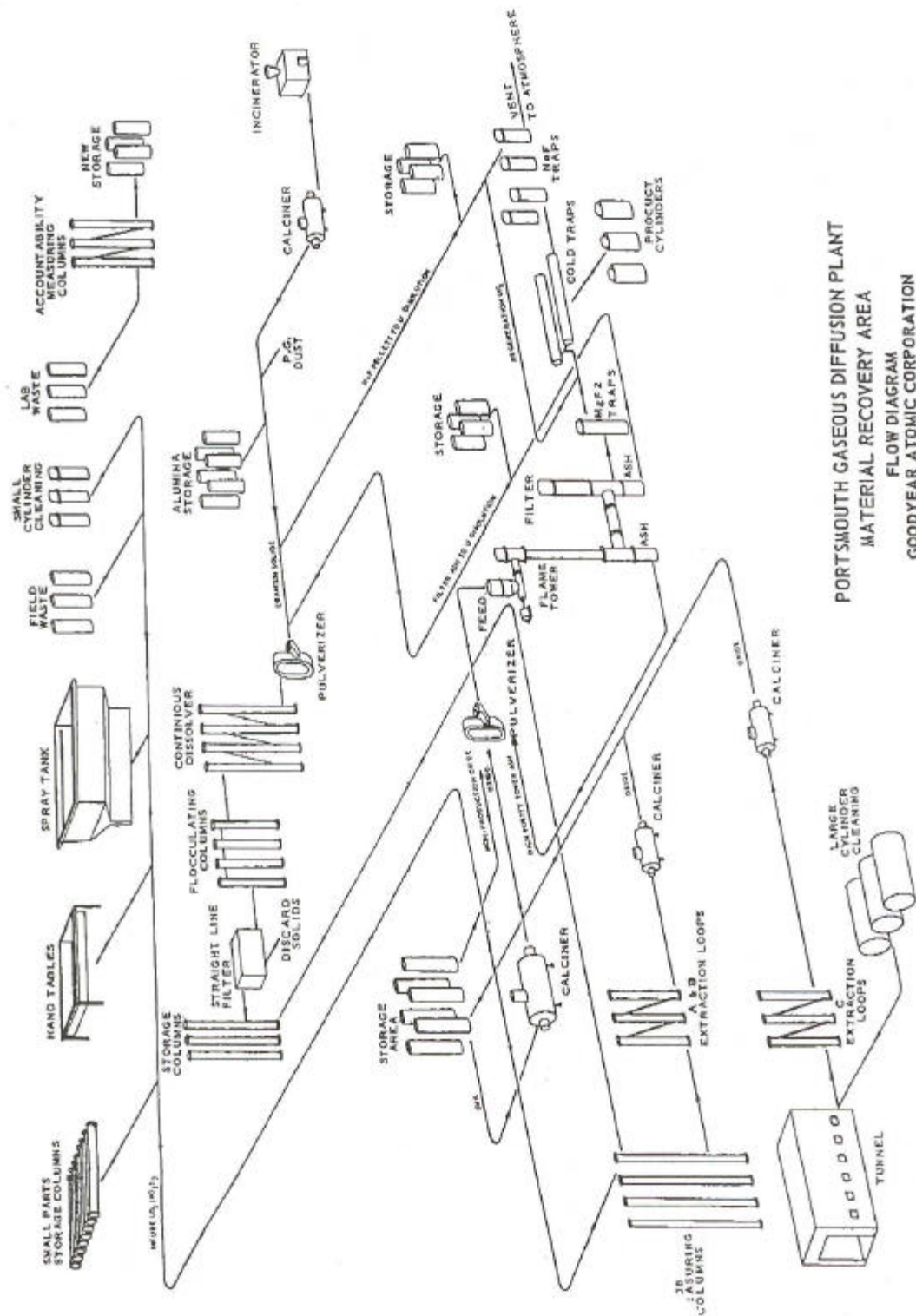
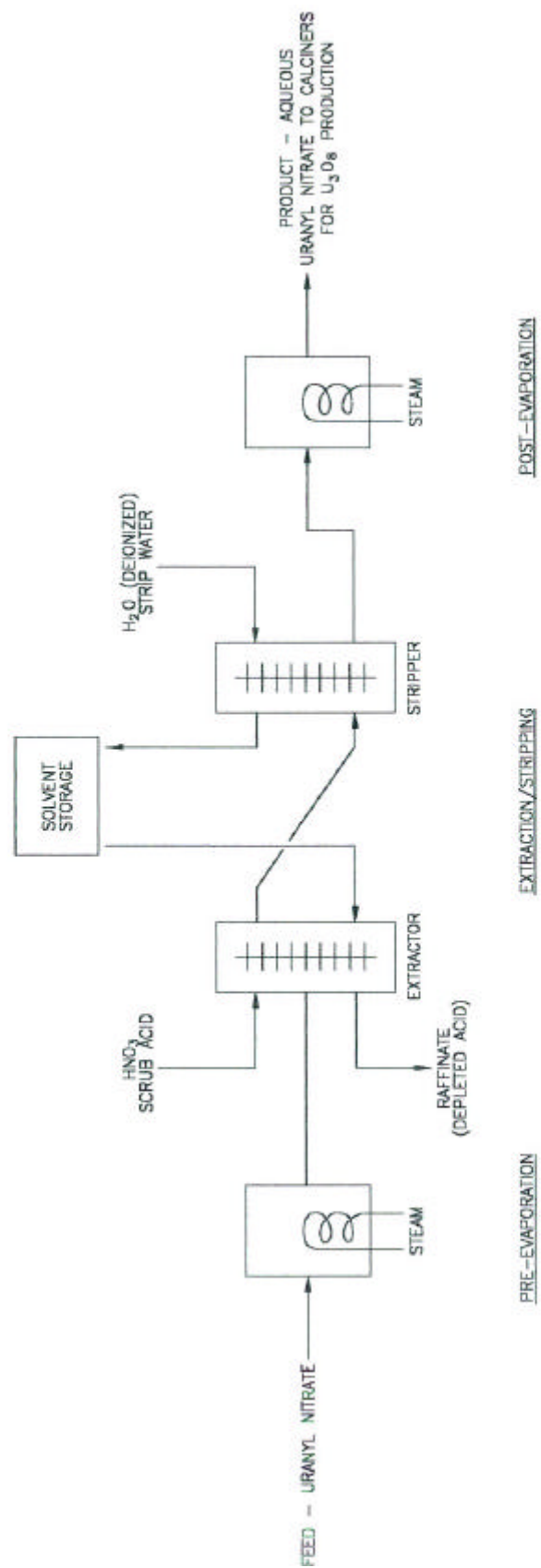
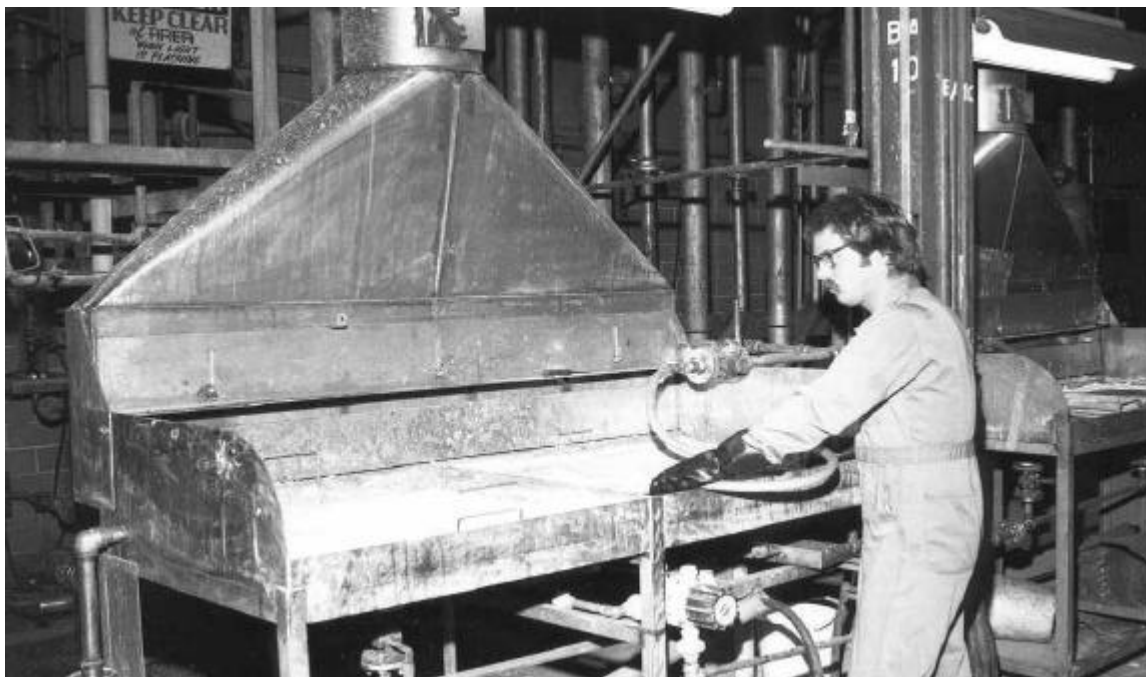


FIGURE 2.2.3.1-2  
SOLUTION RECOVERY PROCESS



SOLVENT: 20% TRIBUTYLPHOSPHATE (C<sub>4</sub>H<sub>9</sub>)<sub>3</sub>PO<sub>4</sub>  
80% VARSOL (PETROLEUM DISTILLATE - KEROSENE)



Chemical Operator Cleaning up UNH Solution at a Filter Table

The  $^{99}\text{Tc}$  appears to take the opposite course, being highly soluble in both nitric acid and water, and forming pertechnetic acid ( $\text{HTcO}_4$ ) (Ref. 10). This acid, however, is not stripped from the solvent and remains in the waste raffinate. While quantitative data could not be found, the literature suggests that essentially all  $^{99}\text{Tc}$  enters the raffinate stream and that has been the assumption for this study.

Concentration of  $^{99}\text{Tc}$ , therefore, takes place in solution recovery in the form of the sludges and precipitates formed from raffinate treatment.

Treatment of the  $^{99}\text{Tc}$  containing raffinate has evolved throughout the plant's history from: (1) rudimentary pH adjustment and discharge to the X-701B/east drainage ditch/Scioto River to (2) secondary pH adjustment and retention at the X-701B/east drainage ditch/Scioto River to (3) today's configuration of extensive treatment via (a) heavy metals precipitation; (b) technetium ion exchange; and (c) biodenitrification with discharge only to a permitted waste water treatment facility.

Concentration of  $^{99}\text{Tc}$  has occurred in: (1) soils surrounding the settling pond and drainage ditch (pre FY 1972), (2) sludges/precipitates occurring from neutralization with lime (FY 1972-FY 1984), and (3) heavy metals neutralization sludge and spent ion exchange resins (since FY 1984)

### 2.2.3.2 Material Flowsheet

Uranium-bearing material forms and flows into and out of the uranium recovery process, (including oxide generation) are shown as Figure 2.2.3.2-1.

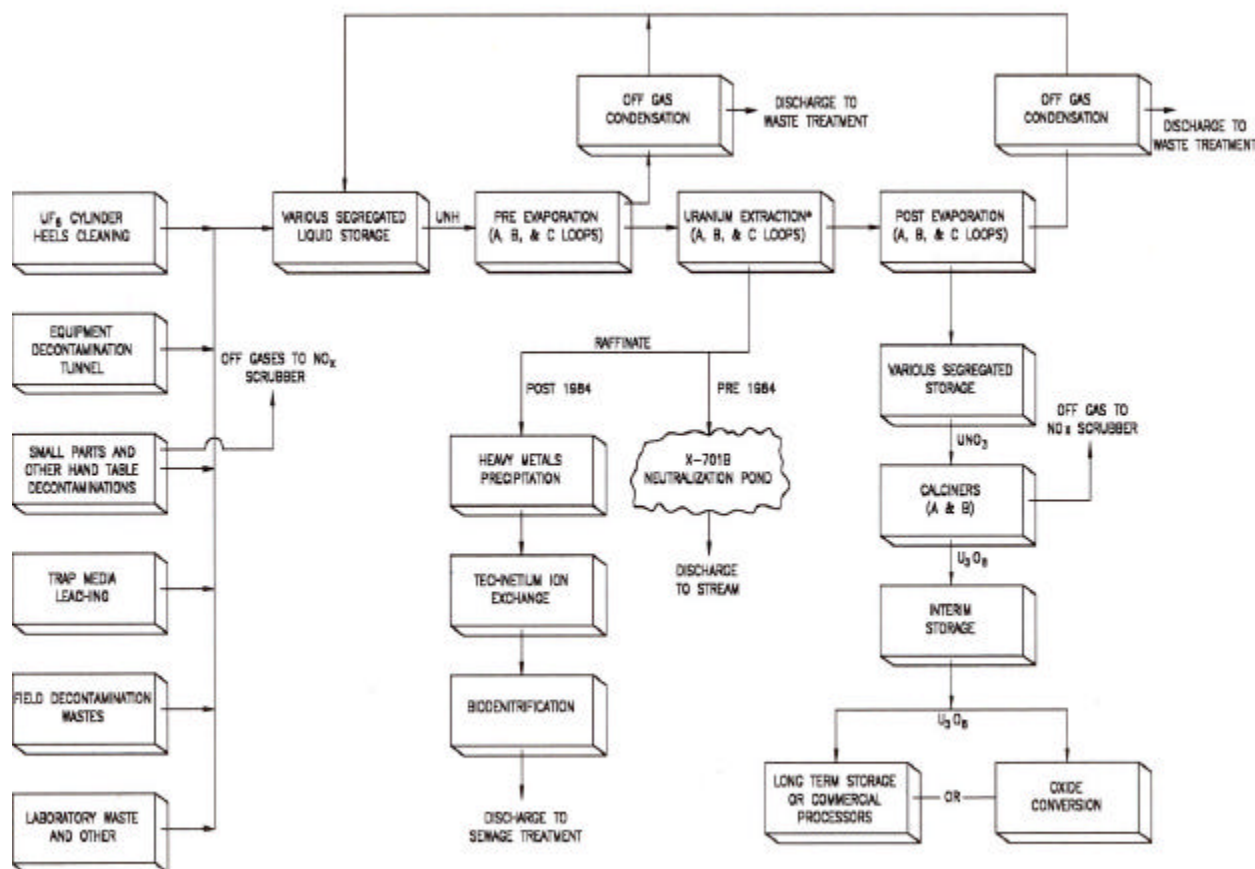
Measurement of these streams has been an uneven process throughout the years covered by this study. Table 2.2.3.2-1 presents the oxide output flows to the extent that the information could be located in plant records (Ref. 11).

Of principal concern with this uranium recovery facility is the  $^{99}\text{Tc}$  content of the raffinate waste stream. Other than spot samples taken during process upset and discovery periods, representative sample data for operating periods can not be found.



Figure 2.2.3.2-1

### Uranium Recovery



\* TRIBUTYL PHOSPHATE AND PETROLEUM DISTILLATE COUNTER CURRENT EXTRACTION PROCESS.

When  $^{99}\text{Tc}$  was first detected in the X-701B (FY 1975), extensive sampling to quantify the  $^{99}\text{Tc}$  constituent was undertaken. As part of the environmental sampling program, monitoring of this stream has continued unabated ever since. Amounts of  $^{99}\text{Tc}$  in this flow are shown in Table 2.5-1 (Ref. 12).

It is postulated that the best quantification of  $^{99}\text{Tc}$  in this waste consists of the characterization studies performed on the sludges dredged from the X-701B. This environmental remediation captured a high percentage of precipitated  $^{99}\text{Tc}$  compounds that were deposited prior to ceasing use of the X-701B facility in FY 1983. Three hundred ninety-one grams (g) of  $^{99}\text{Tc}$  were measured in approximately 2.5 million pounds of sludge. Considering that this sludge contained 1,652 kgU,  $^{99}\text{Tc}$  was concentrated here to 237 ppm on a uranium basis. Adding this to the 1,024 g for years prior to FY 1983 (Table 2.5-1) gives an estimate of 1,415 grams as the total  $^{99}\text{Tc}$  emanating from the raffinates of uranium recovery. It should be noted that in addition to  $^{99}\text{Tc}$ , 0.03 and 3.3 grams of Pu and Np respectively, were measured in the X-701B sludge when it was characterized for LLW disposal. This serves to confirm the imperfect performance of the solution recovery process or the possibility that all TRU-containing materials were not processed through recovery. These sludges are currently being shipped to Envirocare of Utah, Inc.

Table 2.2.3.2-1

## Uranium Recovery Production Data Summary

| Fiscal Year  | Oxide kgU Produced        | Fiscal Year | Oxide kgU Produced |
|--------------|---------------------------|-------------|--------------------|
| 1956         | 726                       | 1978        | 1105               |
| 1957         | 1313                      | 1979        | 1758               |
| 1958         | 1233                      | 1980        | 311                |
| 1959         | 1799                      | 1981        | 1240               |
| 1960         | 2896<br>(Peak Production) | 1982        | 891                |
| 1961         | 1559                      | 1983        | 1127               |
| 1962         | 2032                      | 1984        | 888                |
| 1963         | 526                       | 1985        | 592                |
| 1964         | 493                       | 1986        | 926                |
| 1965         | 640                       | 1987        | 1263               |
| 1966         | 508                       | 1988        | 1330               |
| 1967         | 581                       | 1989        | 561                |
| 1968         | 537                       | 1990        | 640                |
| 1969         | 713                       | 1991        | 641                |
| 1970         | 875                       | 1992        | 218                |
| 1971         | 1745                      | 1993        | 167                |
| 1972         | 863                       | 1994        | 299                |
| 1973         | 585                       | 1995        | 125                |
| 1974         | 447                       | 1996        | 328                |
| 1975         | 773                       | 1997        | 456                |
| 1976         | 1020*                     | 1998        | 333                |
| 1977         | 1069                      | 1999        | 125**              |
| <b>TOTAL</b> |                           |             | <b>38,257</b>      |

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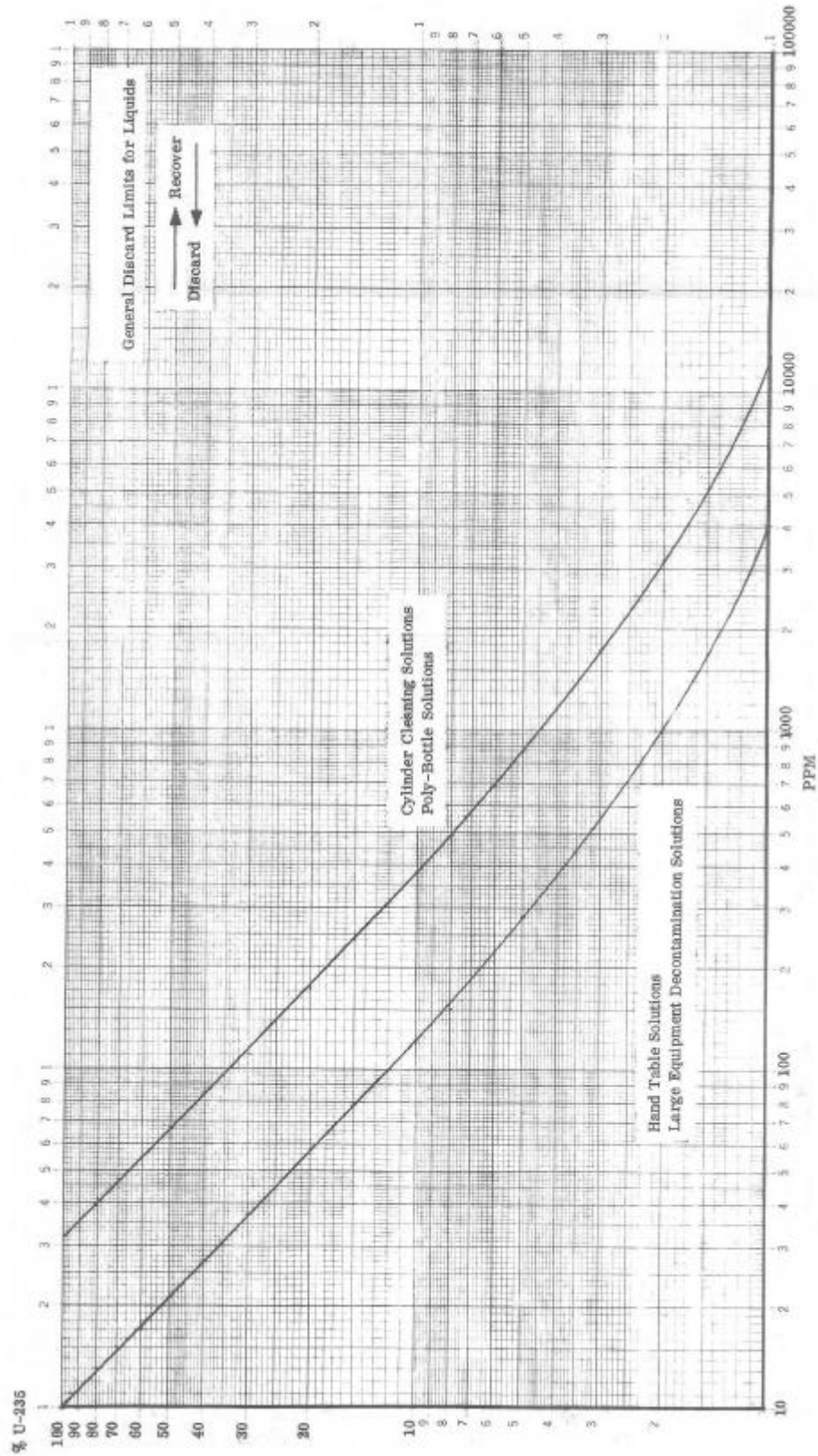
\*15 months

\*\*through March 1999

### 2.2.3.3. Feed Specifications

Go-NoGo criteria for feed to uranium recovery consisted in the past of uranium content (ppm) and enrichment (%  $^{235}\text{U}$ ) nomograms derived from the value of the material and the cost to recover the uranium. A typical example of this is given as Figure 2.2.3.3-1 Discard Criteria for Solutions (Ref. 13). Materials not meeting the reclaim criteria were usually dumped to the X-701B through building drains. No record can be found of consideration of either TRU or FP concentrations as a criteria for feeding. It is suspected that most input streams were never measured for either TRU or FP. Current day operations also do not control inputs. Discharge through building drains was discontinued circa 1984.

Figure 2.2.3.3-1



#### 2.2.3.4. Product Specifications

Product from uranium recovery consists of fully dried and denitrated  $U_3O_8$ . No attempt was made to control purity, TRU/FP content, etc. Levels of denitrification and dehydration are controlled by feed rate to the rotary kiln to produce a product at a prescribed color and dryness based on experience.

#### 2.2.3.5. Operating History



X-701B Holding Pond

The solution recovery system was turned over to GAT on August 3, 1955 and began producing production quantities of  $U_3O_8$  during late FY 1956. While many improvements for operational efficiency and safety (especially nuclear criticality safety) have been made over the life of the facility, the primary process of solvent extraction/stripping and calcination have remained unchanged. Decontamination chemicals (boric acid, sodium carbonate, citric acid, sodium bisulfate, etc.) have been introduced to enhance decontamination efficiency and/or adopt more environmentally friendly technologies during the precursor steps in the process. Major modifications or operational changes to the effluent (raffinate) treatment occurred as follows:

1. In 1972, the precipitation of uranium in the X-701B was enhanced by the addition of facilities to feed slaked lime and photoelectrolyte proportional to the influent rate. This raised the influent stream pH to about 8 in order to promote precipitation of uranium. (This would have also enhanced the precipitation of Np and Pu and some  $^{99}Tc$  compounds).
2. Once or twice each year, the pond was dredged or sludge pumped to containment ponds elevated above and alongside the settling pond.
3. The east containment pond held the dredgings from 1973 through 1980. The west containment pond held the dredgings from 1980 to 1984.
4. During 1984, the discharge of raffinate to X-701B was permanently suspended with the startup of the heavy metals precipitation process (X-705),  $^{99}Tc$  ion exchange process (X-705), and the biode-nitrification process (X-700).

5. The X-701B and the two companion containment ponds were dredged with their contents and about one foot of bottom soil being containerized and characterized for disposal as LLW. Shipment of these soils to Envirocare of Utah, Inc. is in progress.

The peak year of operation occurred in FY 1960 when this facility processed nearly 8 MTU probably predominately from solutions generated from the first process change-out program. Average production rates were approximately 0.9 MTU per year over the life of the facility. Total production of  $U_3O_8$  for the approximately 44 years of operation through March 1999 was about 38.2 MTU. Highlights of the operation include sustained high levels of throughput during the period FY 1974 through FY 1983, much of which was in support of the second diffusion process change-out known as the Cascade Improvement Program/Cascade Upgrading Program (CIP/CUP).

#### **2.2.3.6 Current Status**

Uranium recovery remains in service to support ongoing efforts of USEC to maintain the gaseous diffusion plant operation. Extensive revisions to nuclear criticality safety driven procedures have been made and other changes have been initiated in support of Technical Safety Requirements of the NRC-USEC license.

### **2.2.4 Oxide Conversion**

#### **2.2.4.1 Plant Description**

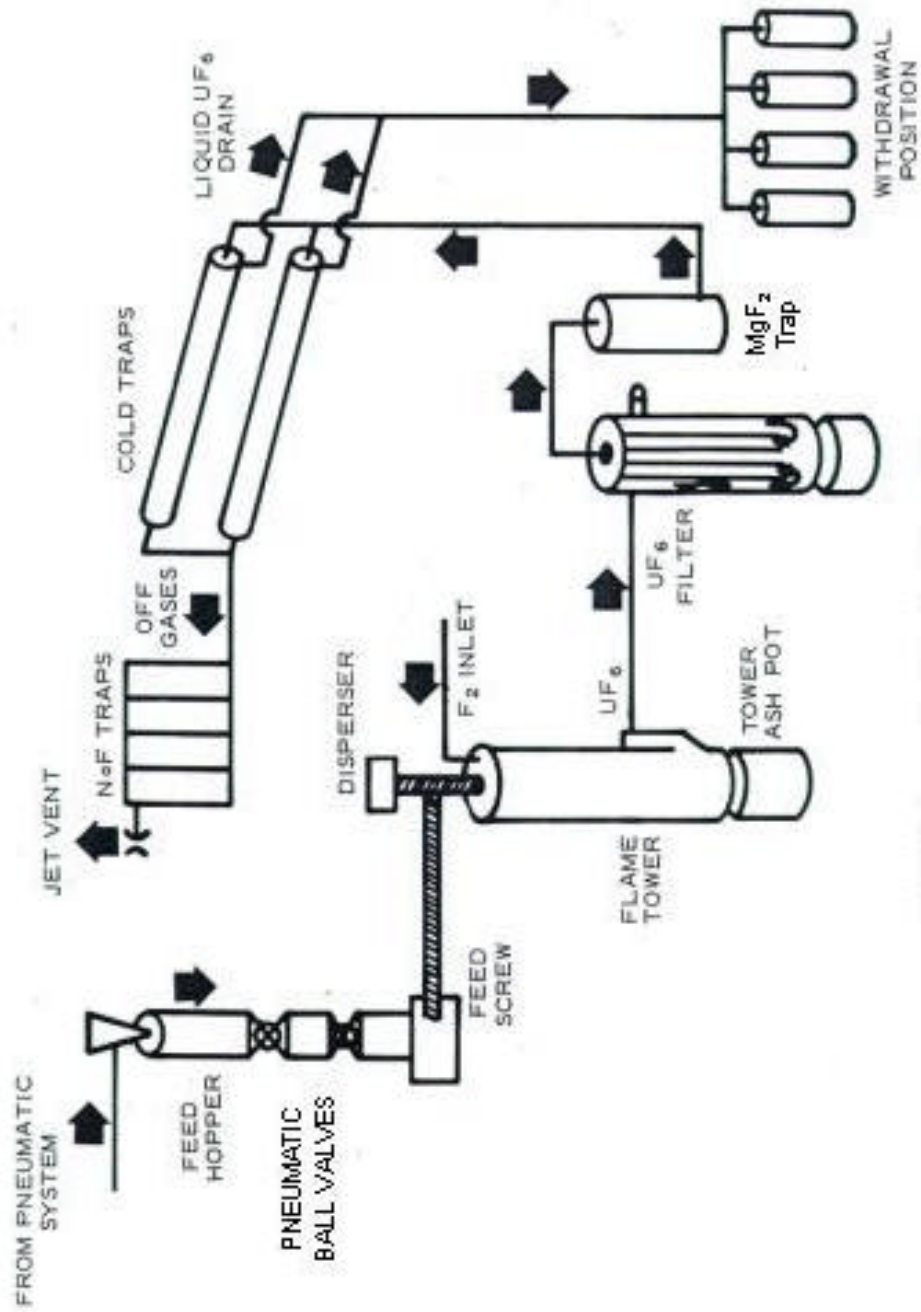
The Oxide Conversion Facility is located in “E”, “F”, and “H” areas of X-705. It was supplied as part of the original plant equipment complement and was designed to be an integral part of the uranium recovery process (see Figure 2.2.3.1-1). Over time, this facility developed a somewhat unique mission and set of operating streams and as such is dealt with separately for this report.

To be used as feed to the gaseous diffusion process, uranium oxides originating from a variety of operations (both on-site and off) must be converted (fluorinated) to  $UF_6$ . This process of fluorination was originally performed at PORTS in a system of three parallel, horizontal, screw-fed, stirred-bed reactors. In these reactors,  $U_3O_8$  was reacted with fluorine to form  $UF_6$  which was cold trapped in chilled 5” cylinders. Little is known about this early system other than the fact that it was unreliable and had inadequate production capabilities. In 1959 the stirred-bed reactors were replaced with a site-developed 4” diameter open flame tower using direct fluorination. Stated capacity of the system was 7,200 kgU/yr output as  $UF_6$ .

In mid-1965, AEC approved a project to upgrade the Oxide Conversion Facility to achieve a production capability of 20,000 kgU/yr as  $UF_6$  in support of its designation of PORTS as the processor of uranium scrap >10% enrichment for the DOE complex. The upgrade was accomplished in 1966 and 1967 and consisted of installing a significantly improved 5” flame tower, an improved feed system, an improved product filtering and withdrawal system, and an improved off-gas treatment and monitoring system. A pneumatic conveying system was installed to facilitate movement of the oxides and ashes. Where release of dusty powders might be expected, gloveboxes were installed. A pictorial diagram of the fluorination process is shown as Figure 2.2.4.1-1. A more detailed description of the process and its history are available in Ref. 14.

The process worked as follows: Oxides were fed into the top of the flame tower where they fell by gravity through a fluorine jet. Unburned (unreacted) oxide and some impurities would fall into the ash pot below the tower where, on a batch basis, the ash would normally be removed, ground and refeed to the top of the tower.  $UF_6$  generated in the tower would pass through a sintered metal filter to remove particulates and then through a  $MgF_2$  trap for sorption of certain impurities.

Figure 2.2.4.1-1

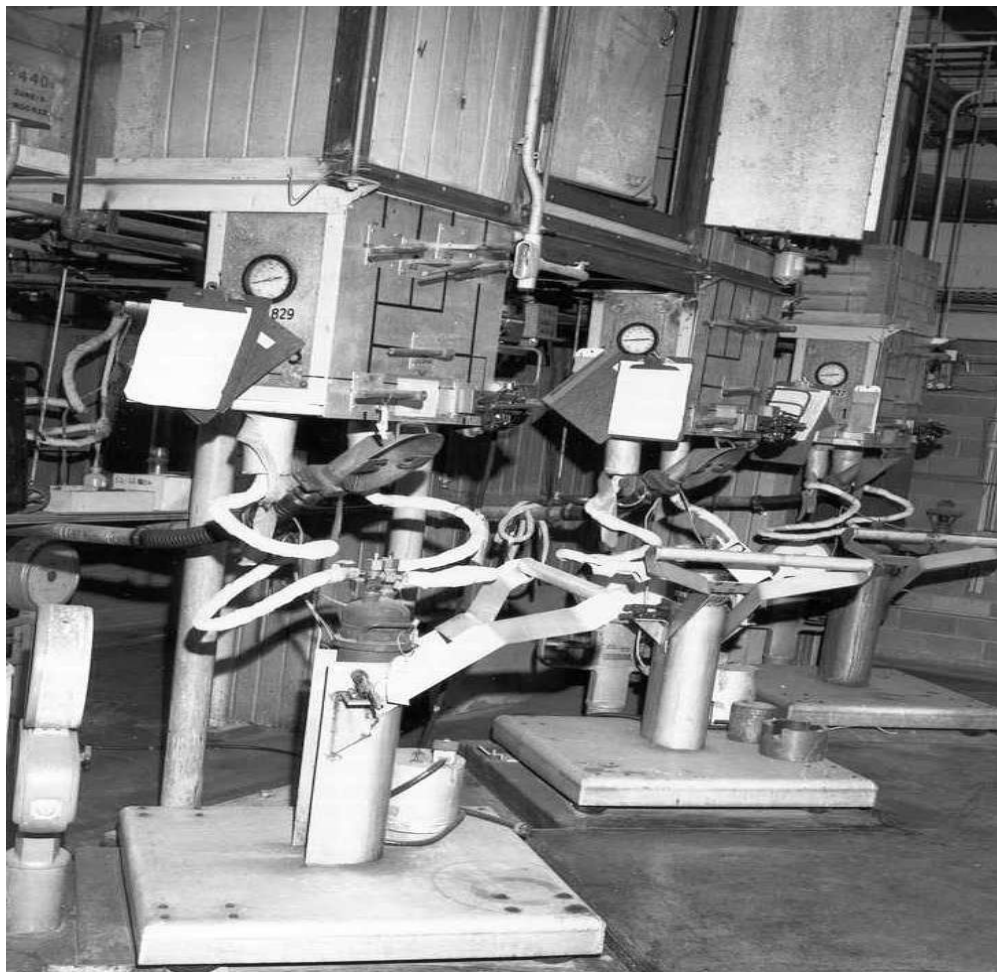


Oxide Conversion System Flow Diagram



Gaseous  $\text{UF}_6$  was condensed in cold traps with liquid  $\text{UF}_6$  drained by gravity to 5", 8", 12", or 2-1/2-ton cylinders. Off gases from the cold traps were passed through NaF traps to remove remaining minor amounts of  $\text{UF}_6$  prior to ejection through a monitored vent.

In theory, TRU/FP's fed to this process take the following paths: Pu oxide formed gaseous  $\text{PuF}_6$  in the flame tower should quickly become solid  $\text{PuF}_4$  due to spontaneous dissociation and fall out in the tower ash or be filtered by the sintered metal filter (filter ash). Fluorinated Np will be sorbed on the  $\text{MgF}_2$  trap. Technetium is generally not expected in the feed. If it was present, it would be easily fluorinated, sorbed on the  $\text{MgF}_2$  or NaF traps or be vented. Based on these assumptions, Pu should concentrate in the tower ash and filter ash. Np should concentrate on  $\text{MgF}_2$ .



Oxide Conversion Cylinder Fill Positions

The  $^{99}\text{Tc}$  will not be a factor except in the vent. Product cylinders should contain only trace amounts of TRU/FP's. In reality, Pu and Np both are found to be concentrated (based on uranium feed) from time to time in tower and filter ashes,  $\text{MgF}_2$  trapping media, and in  $\text{UF}_6$  product cylinders. Based on sample analysis data (Ref. 15), it appears that concentrations of constituents of concern in the waste and product streams are highly dependent on the degree of ash recycle, the in-service life of the trap media, as well as the constituent concentrations in the oxide feed. Perhaps operating parameters such as tower temperature also influence these concentrations. While a detailed parametric analysis of all variables could not be found, an analysis based on TRU sample data from a May 1976 run involving TRU material received from NLO gives insight into the distribution of constituents. Table 2.2.4.1-1 shows some of this data.

**Table 2.2.4.1-1**

**Material Balance of Transuranics in Oxide Conversion During May 1976**

| Process Step                 | Uranium<br>(kgU) | <sup>237</sup> Np<br>(g) | <sup>238</sup> Pu<br>(ug) | <sup>239</sup> Pu<br>(ug) | <sup>241</sup> Pu<br>(ug) |
|------------------------------|------------------|--------------------------|---------------------------|---------------------------|---------------------------|
| Input Oxide                  | 3241             | 1.133                    | 96.3                      | 13510                     | 231                       |
| Output                       |                  |                          |                           |                           |                           |
| Solution                     | 6.7              | 0.003405                 | 0.5                       | 196                       | 0.7                       |
| Filter Ash                   | 0.9              | 0.1419                   | 21.2                      | 5152                      | 8.8                       |
| Tower Ash                    | 14.8             | 0.0112                   | 2.2                       | 344                       | 3.1                       |
| Oxide                        | 5.1              | 0.0034                   | 0.9                       | 84                        | 1.3                       |
| Sodium Fluoride (NaF)        | 3.5              | 0.0118                   | 0.1                       | 34                        | -0-                       |
| Total Output                 | 31.0             | 0.1717                   | 24.9                      | 5810                      | 13.9                      |
| % Output/Input               | 0.96             | 15.2                     | 25.8                      | 43.0                      | 6.0                       |
| % In product Cylinders (max) | 99               | 84.8                     | 74.2                      | 57.0                      | 94.0                      |

From this limited data (UF<sub>6</sub> output and MgF<sub>2</sub> holdup were not analyzed), it can be seen that as much as 85% of the <sup>237</sup>Np and 57% of the <sup>239</sup>Pu may be present in the UF<sub>6</sub> product cylinders. Of that not in the UF<sub>6</sub> product, the filter ash is seen to contain the majority of the TRU constituents (12.5% of <sup>237</sup>Np and 38% of <sup>239</sup>Pu). The concentrating effect of these streams relative to input uranium concentration is illustrated in Table 2.2.4.1-2.

**Table 2.2.4.1-2**

**TRU Concentration Factors from Table 2.2.4.1-1**

| Process Step | <sup>237</sup> Np<br>(g/gU) | Concentration<br>Factor ( <sup>237</sup> Np) | <sup>239</sup> Pu<br>(g/gU) | Concentration<br>Factor ( <sup>239</sup> Pu) |
|--------------|-----------------------------|--|-----------------------------|--|
| Input Oxide  | 3.5 x 10 <sup>-7</sup>      | Base   | 4.2 x 10 <sup>-9</sup>      | Base   |
| Filter Ash   | 1.6 x 10 <sup>-4</sup>      | 451  | 5.7 x 10 <sup>-6</sup>      | 1375   |
| Tower Ash    | 7.6 x 10 <sup>-7</sup>      | 2.2  | 2.3 x 10 <sup>-8</sup>      | 5.6  |
| NaF          | 3.4 x 10 <sup>-6</sup>      | 9.7  | 9.7 x 10 <sup>-9</sup>      | 2.3  |

The filter ash for this case is seen to have concentrated the <sup>237</sup>Np by 451 times and the <sup>239</sup>Pu by 1,375 times relative to the levels in the uranium fed to the facility.

Analysis of filter ash remaining on site and analyzed as part of the HEU removal characterization studies showed average concentrations of: 1.9 x 10<sup>-4</sup> and 3.3 x 10<sup>-6</sup> g/gU, respectively, and peak concentrations of 3.9 x 10<sup>-4</sup> and 5.8 x 10<sup>-6</sup> g/gU, respectively, of <sup>237</sup>Np and <sup>239</sup>Pu which are reasonably close to those in the NLO material balance study.

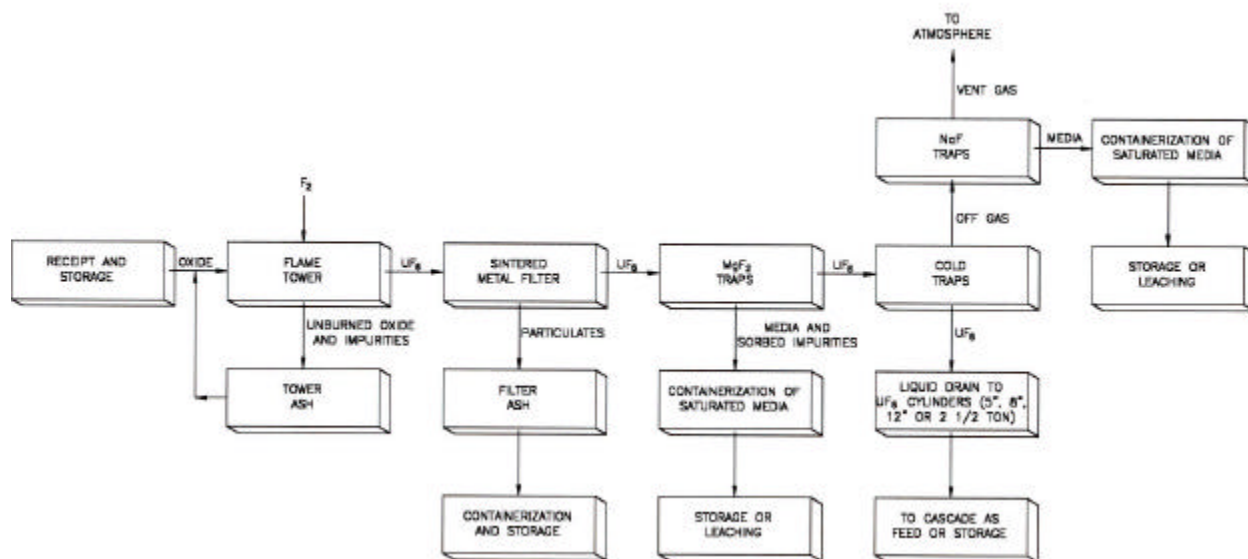
As a result of these data, the oxide conversion process is considered as removing at least 15% of <sup>237</sup>Np and 43% of <sup>239</sup>Pu.

### 2.2.4.2. Material Flowsheet

Uranium bearing material flows through the oxide conversion process are shown in Figure 2.2.4.2-1.

Figure 2.2.4.2-1

Oxide Conversion Flow Sheet



Accurate quantification records of these streams has been an uneven process throughout the life of the facility. Table 2.2.4.2-1 presents the UF<sub>6</sub> produced from oxide based on information available in plant records (Ref. 11).

Table 2.2.4.2-1

Oxide Conversion Production Data Summary

| Fiscal Year   | UF <sub>6</sub> Produced<br>kgU | Fiscal Year             | UF <sub>6</sub> Produced<br>kgU |
|---|---------------------------------|-------------------------|---------------------------------|
| 1958  | 2,795                           | 1967                    | 0                               |
| 1959  | 1,425                           | 1968                    | 6,622                           |
| 1960  | 887                             | 1969                    | 20,257                          |
| 1961  | 1,170                           | 1970                    | 7,712                           |
| 1962  | 866                             | 1971                    | 1,580                           |
| 1963  | 497                             | 1972                    | 48,550                          |
| 1964  | 978                             | 1973                    | 51,344                          |
| 1965  | 4,085                           | 1974                    | 16,650                          |
| 1966  | 4,995                           | 1975                    | 22,915                          |
| <b>Total Old System</b>                                     | <b>17,698</b>                   | 1976 (5 Quarters)       | 28,710                          |
|   |                                 | 1977                    | 7,806                           |
|   |                                 | 1978                    | 3,252                           |
|   |                                 | <b>Total New System</b> | <b>215,398</b>                  |
| <b>Total Life of Facility 233,096 kgU as UF<sub>6</sub></b> |                                 |                         |                                 |

Of principal concern with this facility is the TRU content of the feed stocks and their concentration in the product and waste streams.

Significant amounts of TRU materials were known to have been processed during the two periods shown in Table 2.2.4.2-2 (Ref. 16).

**Table 2.2.4.2-2**

| Period of Operation | Quantity Processed kgU | Origin of Material | Probable Total Alpha from TRU dpm/gU |
|---------------------|------------------------|--------------------|--------------------------------------|
| Jan.-Feb. 1974      | 1373                   | ICPP               | <2000                                |
| Jan. & May 1976     | 4214                   | NLO                | <3000                                |

There is some indication that as much as 3.7 MTU of recycled depleted oxide ( $\text{UO}_2$  and  $\text{UO}_3$ ) from the Oak Ridge K-25 site may have been processed in the 1958 – 1961 time frame. Shipping records for this material specify that it was to be used for research and development purposes. It is believed that this material was used during the development of the fluorination tower (FluorOx) in the X-760 Chemical Engineering Building or during unsuccessful development efforts for oxide pelletization. Since this material was depleted and of extremely low value compared to HE/VHE materials it seems unlikely that the limited oxide conversion capacity in the X-705 would have been used to process this material. It would have amounted to approximately 75% of the total production of the Oxide Conversion Facility during the 1958 – 1961 time period. The provenance of this material is unknown and remains in the disposition uncertain category.

There was a significant amount of TRU/FP (6847 kgU) as UNH calcined to  $\text{U}_3\text{O}_8$  in the oxide conversion calciner during March – December 1977. This material was never converted to  $\text{UF}_6$  at PORTS and was later shipped to NLO.

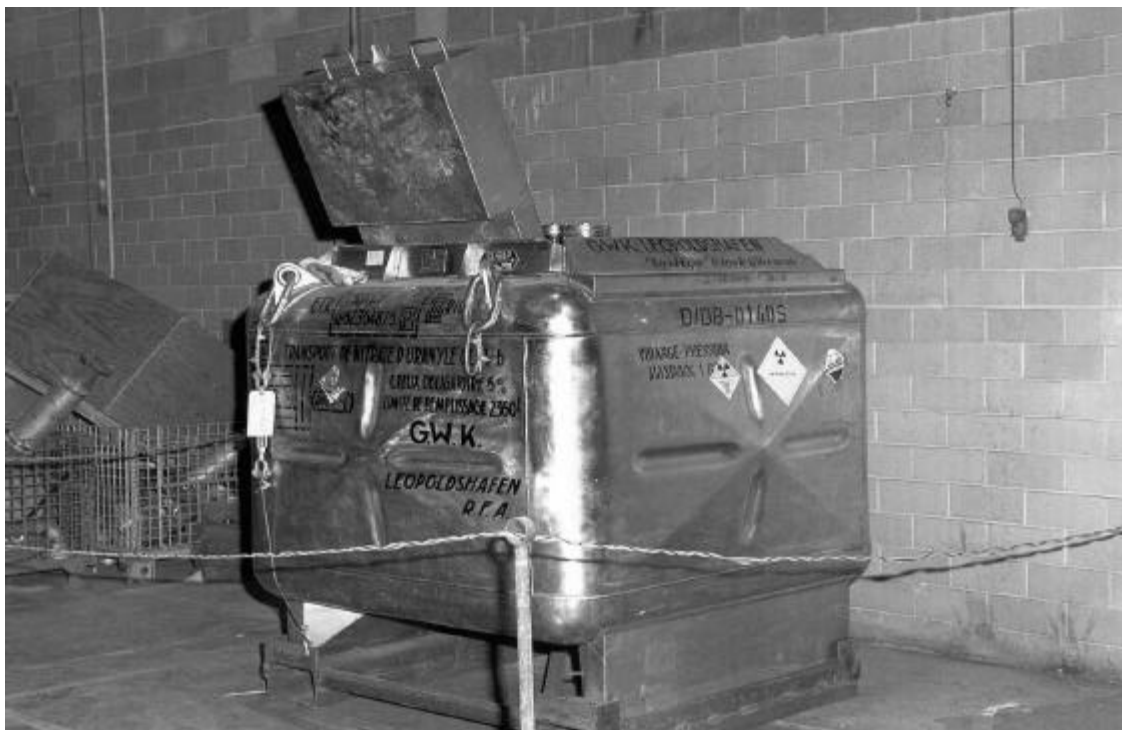
Side streams from the process were generally treated as follows:

1. Tower Ash: Recycle to tower as feed as soon as possible after generation.
2. Filter Ash: Some unknown (possibly minor) amounts were digested at uranium recovery to create  $\text{U}_3\text{O}_8$  for a second attempt at conversion to  $\text{UF}_6$ . The filter ash that remains on-site is stored in the X-326 “L” Cage. Partial contents of the amount remaining are shown in Table 2.2.4.2-3.
3. Magnesium Fluoride: There are indications that some of this material may have been leached at uranium recovery. There remains on site a quantity of  $\text{MgF}_2$  that, in part, may have been generated from oxide conversion. Quantification of this stream is to be determined.

**Table 2.2.4.2-3**

**Filter Ash (Partial Contents) Stored in X-326 “L” Cage**

| Constituent       | Quantity (g) |
|-------------------|--------------|
| Filter Ash        | 40,725       |
| Uranium           | 3,007        |
| $^{235}\text{U}$  | 1,233        |
| $^{237}\text{Np}$ | 0.563        |
| $^{239}\text{Pu}$ | 0.010        |



German "CUBE" UNH Shipping container

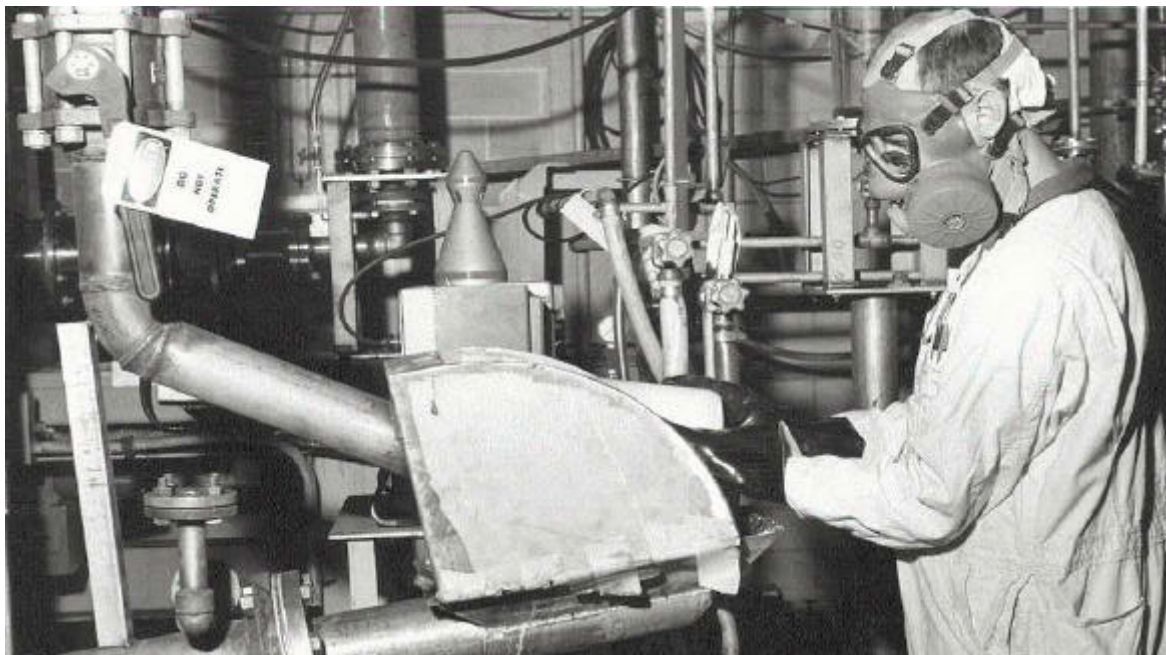
4. Sodium Fluoride: The facility was designed to regenerate the NaF traps through in-place heating and vaporization of sorbed uranium. Regenerated  $UF_6$  would be introduced into the inlet of the cold traps, condensed, and drained into  $UF_6$  cylinders. After several regeneration cycles, the NaF loses its absorption capability and must be replaced. This depleted/removed trap media may contain TRU. Also, whenever inventory of the system was required, usually monthly, the NaF was removed and the uranium leached, measured, and reintroduced into the uranium recovery process, if economical. The TRU on the media may accompany the media or be leached with the uranium. Depleted/leached media was containerized, the cans placed into site-prepared wooden boxes with void spaces filled with lime, and the boxes were buried at the X-749A landfill.

#### 2.2.4.3. Feed Specifications

Feed to oxide conversion originated from both on-site and off-site sources. A specification for material originating from on-site could not be found. It is probable that if the oxide was of an enrichment level corresponding to a planned production campaign, it was considered an acceptable feed stock. The facility was operated to minimize assay mixing losses and, as such, quantities of materials at desired enrichments became the criteria for feeding. It is probable that materials originating on-site were seldom, if ever, fully analyzed except for uranium and  $^{235}U$  content. Materials originating off-site were required to satisfy: "Feed Specification for U-235 Enriched Uranium Returned to AEC" (Ref. 17).

Since the  $UF_6$  produced in oxide conversion was destined to be fed to the diffusion cascade, specific limits were placed on many parameters (usually metals) that would result in out-of-specification product from the diffusion cascade (nuclear poisons, etc.). Total TRU limits were expressed as total alpha activity from TRU and from the earliest days of the scrap returns program were set at 1,500 dpm/gU. Deviations from the acceptance specification were numerous throughout the life of the scrap program. These deviations were primarily due to excessive amounts of various non-TRU metals. Numerous letters accompany the transaction records prescribing minimal monetary penalties in an effort to indemnify AEC for cost of dealing with off-specification materials. No examples, however,

of deviations for TRU with accompanying penalties were found. There was substantial correspondence between AEC site managers or operating contractors discussing capabilities to accept various forms and levels of TRU. Materials from NLO were knowingly accepted in the FY 1975 - FY 1976 timeframe with gross alpha from TRU levels up to 3,000 dpm (Ref. 18).



Chemical Operator Introducing UNH Crystals to Dissolver Ahead of Calciner

#### **2.2.4.4 Product Specification**

No formal specification existed covering the production of  $UF_6$  from oxide conversion. As stated earlier, impurities in product became an issue in-as-much as they would impact the cascade and its product. Since this  $UF_6$  would ultimately be blended in the cascade with a large amount of other feed stocks, timing and scheduling its feeding to the cascade could frequently mitigate any adverse contaminants. Materials unable to be fed to the cascade were produced at oxide conversion and were still present at the suspension of HEU production in 1991.

#### **2.2.4.5 Operating History**

A significant event timeline and a detailed explanation of oxide conversion's history are available in Reference 14. In summary, the facility operated from 1957 to 1978. It produced about 233 MTU of  $UF_6$  from materials originating from at least 47 feed sources. It is known that 5.6 MTU of feed contained TRU's. Operations were terminated when it became apparent that the existing facility could not meet current standards for containment as manifested in high levels of airborne contamination. Additional efforts to modernize, renovate, or replace the facility were terminated for the last time in July 1981 when cost estimates to provide this capability at PORTS indicated prohibitively high costs.

#### **2.2.4.6 Current Status**

Attention to this facility since shutdown has been limited to custodial activities that assure it remains safe and secure. The facility is locked to prevent spread of transferable contamination. Minor amounts of contamination remain internal to the system. There currently are no known funded plans for Decontamination and Decommissioning (D&D) of this facility.



### 2.2.5 Other Uranium Handling Facilities

Other uranium handling facilities are X-710 Technical Services Building (laboratory), X-760 Chemical Engineering Building, X-744G Bulk Storage Warehouse (uranium storage warehouse), X-345 Special Nuclear Materials (SNM) Storage Building, and X-745 Cylinder Storage Yards.

#### 1. X-710 Technical Services Building

The X-710 laboratory was part of the original plant facilities. With respect to uranium, the primary analytical capabilities of the laboratories are:

- a. Isotopic analysis (mass spectrometry) for feed and product;
- b. Uranium purity (Davies Grey, wet chemistry);
- c. Metallic impurities (ICP, spectral analysis);
- d. Radiological analysis (radiochemistry, alpha, beta, gamma, counting); and
- e. Sample preparation and waste handling processes for all of the above.

For this report, quantities processed were considered small with minimal likelihood of concentration.

Development of processes and procedures for plant use has been an integral part of this facility's mission for most of its life. There were a myriad of experiments that involved TRU and FP that potentially separated or concentrated these constituents. These development activities involved minor/trace amounts of these elements/isotopes. For this report, no attempt to quantify these is made. The likelihood of relevance to the site mass balance is low, but not zero.

#### 2. X-760 Chemical Engineering Building

The X-760 Chemical Engineering Building had, as its mission, the pilot-scale development work on new chemical processes prior to or in aid of plant deployment. This facility had a small process laboratory, a small machine/fabrication shop, a worker change/shower area, and essentially all plantsite utilities. Early development projects, including decontamination process experiments, boiling freon heat exchanger experiments,  $\text{UF}_6$  heating studies, uranium oxide pelletizing experiments, freon drying tests, and controlled  $\text{UF}_6$  releases in a sealed environmental chamber, were conducted in this facility. Most relevant for this study is the prototype development work on what is referred to in reports as the fluorox process. This (what appears to be) fluorination tower may have been prototypical or a developmental aid either for the  $\text{UF}_6$  feed manufacturing facility (X-344) or the oxide conversion fluorination system (X-705). Records show that 0.86 MTU of  $\text{UF}_4$ , 0.4 MTU of  $\text{UO}_2$ , and 3.3 MTU of  $\text{UO}_3$  were received from K-25 in 1957 and classified as recycled uranium for research and development studies. The final disposition of this material could not be determined. It is likely that any materials converted to  $\text{UF}_6$  were fed to the cascade, and that unconverted materials were converted to  $\text{UF}_6$  during early oxide conversion X-705 operations (probably after 1962). The provenance of the TRU/FP constituents can only be speculated upon. Ash and trapping media disposition records can not be located.

3. X-744G Bulk Storage Warehouse

The X-744G was one of the earliest facilities constructed at PORTS. It was the Peter Kiewit (prime construction contractor) pipe fabrication facility. Here, process piping assemblies were fabricated for cascade and utilities. Interior surfaces of the process piping were also degreased in this facility using organic solvents. Later through most of the early years of plant operation, the building served as the non-UF<sub>6</sub> and small cylinder UF<sub>6</sub> storage area. Overflow materials from X-705 (solutions, UNH, oxides, etc.) were also stored here. Security systems provided protection for HEU materials.

With the advent of the uranium scrap returns program (circa 1966), X-744G was designated as the scrap storage warehouse and central receiving facility. Here oxides, UNH, and small cylinders of UF<sub>6</sub> were off-loaded from transport, with non-UF<sub>6</sub> containers opened and sampled (usually in a hood or glovebox), and placed on shelves or in holders for storage and future conversion at X-705. Materials not meeting acceptance criteria were packaged and shipped from this facility. After cessation of oxide conversion operations, the facility remained in service for storage of all oxides until X-345 was placed in service to store HEU materials. Throughout the life of this facility, no concentrating operations could be determined. There was, however, significant material throughput with opportunity for worker interaction.

This facility currently supports the DOE PORTS Uranium Management Center activities. Quantities of materials containing TRU are being stored here as part of this program. These materials were received after the March 1999 timeframe and, as such, are outside of scope for this report. This facility, over time, has also been the center for other activities on a shared basis such as aluminum smelting, waste sorting, etc. None of these activities are considered as relevant to this report.

4. X-345 Special Nuclear Materials Storage Building

X-345 was constructed, circa 1978, and was basically designed to fill the mission of X-744G for storage of HEU materials but in the more secure environment. An additional mission, added after initial construction, was the sampling of HEU UF<sub>6</sub> small diameter cylinders using autoclaves.

5. X-745 Cylinder Storage Yards

X-745 yards (a through h) are the eight UF<sub>6</sub> cylinder storage yards that exist or have existed at PORTS. These yards, at any one time, constitute the majority of the uranium materials at PORTS and contain UF<sub>6</sub> not in process or not shipped (i.e. feed, product and tails). The depleted UF<sub>6</sub> storage areas are of particular interest due to the buildup of naturally occurring uranium daughter products. Breaching of cylinders due to mechanical failures has occurred. There are no known cases of these breechings occurring in cylinders containing RU.

The repeated filling of UF<sub>6</sub> cylinders that contain RU heels without washing/removing the nonvolatile heel from the cylinder has the effect of concentrating TRU and FP in the heel. The degree to which the constituents concentrate depends primarily on the amounts added with each filling and the fraction removed through feeding. While the exact fraction removed is variable, it is assumed for this study to be as shown in Table 2.2.5-1.

It is known that large numbers of 2-1/2 and 10-ton cylinders were filled with RU off-site (PGDP, ORGDP) and fed at PORTS. These cylinders were either returned empty or filled with non-TRU (tails) and sent back. The potential exists for several such cycles to have been made on a very few cylinders either with RU before they were cleaned, or, they never have been cleaned and the TRU heel remains.

**Table 2.2.5-1****Percentage of TRU/FP Removed from Cylinders During Feeding**

| <b>Constituent</b> | <b>Removal Fraction With Each Feeding</b> |
|--------------------|---|
| <sup>237</sup> Np  | 33% (Refs. 2 and 2a)                      |
| <sup>239</sup> Pu  | 10% (Refs. 2 and 2a)                      |
| <sup>99</sup> Tc   | 90% (Refs. 2a and 19)                     |

PORTS has had a closed system for small diameter (i.e., 5", 8", and 12") cylinder cleaning since the X-705 facility startup in 1957. The capability to clean large (2-1/2, 10, 14-ton) cylinders was established through facility modifications made circa 1970. For this study, no TRU/FP is considered to have entered the PORTS site from cleaning of interplant RU cylinders.

Small diameter cylinders were used at PORTS to contain UF<sub>6</sub> (usually HEU) from X-705 oxide conversion. Specific cylinders were used for this flow and were selected for use based on uranium assay of last use so as to minimize mixing losses with the heel. Since on at least two known occasions (i.e., 1974 and 1976) oxide conversion produced UF<sub>6</sub> from oxides containing elevated TRU levels, the UF<sub>6</sub> produced is considered to contain TRU's. The small diameter cylinders used for this production may have been used several times without cleaning. It is probable that concentrating of <sup>239</sup>Pu and <sup>237</sup>Np took place in these cylinders. Records to corroborate this have not been found, other than records noting the presence of TRU contamination at the facility. There were records of small diameter cylinders being cleaned in about the correct timeframes, but records of specific cylinder numbers cleaned have not been found.

Several small diameter UF<sub>6</sub> cylinders containing HEU RU were received at PORTS from offsite sources {France, NUMEC, Department of International Affairs, Babcock & Wilcox (B&W), and United States Atomic Energy Commission Safeguards and Security (USAEC S&S)}. The majority of this material was only recently fed (circa 1997) to the cascade, and the cylinders with heels were either cleaned in X-705 or Nuclear Fuel Services (NFS). Solutions generated from the recent cleanings in X-705 appear not to have ever been analyzed for TRU. Due to X-705 operational problems, the majority of these solutions (blended down to <5% enrichment) appear to never have been processed and remain in storage. If warranted, they could be sampled and analyzed to ascertain TRU content. The disposition of the cylinders sent to NFS and their solutions are unknown.

### **2.2.6 Intrafacility Flow of TRU/FP Constituents**

Opportunities for cross flow or cross contamination between various streams are worthy of mention. Examples are:

1. Dissolved oxide conversion filter ash reintroduced into uranium recovery;
2. Dissolved ash from personal protective equipment (PPE) and other incinerated burnables, and subsequently, introduced into uranium recovery;
3. X-705 solution complexing to aid recovery of uranium from NaF with the use of alumina, and processing through uranium recovery;

4. Field decontamination solutions originating from TRU contaminated equipment being processed through uranium recovery; and
5. Wastes generated from laboratory operations involving TRU being introduced into uranium recovery.

Any of these intermittent batch operations, as well as perhaps others, could have caused RU to appear at a time other than when RU operations were known to have occurred. The overall effect of these flows would primarily be a slight, but unquantified, increase of chances for personnel exposure.

## 2.3 Activity Summaries (concentrating processes and other site specific issues related to processing and plants)

The Table 2.3-1 summarizes concentrating processes and site specific issues for PORTS.

**Table 2.3-1**  
**Concentrating Processes at PORTS**

| Concentrating Process/Location                                   | Dates of Operation | Comment  |
|--|--------------------|--|
| X-344 UF <sub>4</sub> – UF <sub>6</sub> Tower Ash                | 5/58 – 2/62        | Operation only on non-RU (virgin) material (normal assay)                    |
| X-344 UF <sub>4</sub> – UF <sub>6</sub> Filter Ash               | 5/58 – 2/62        | Operation only on non-RU (virgin) material (normal assay)                    |
| X-705 U <sub>3</sub> O <sub>8</sub> – UF <sub>6</sub> Tower Ash  | 2/57 – 7/77        | TRU's known to be processed Jan./Feb. 1974 – ICPP, and Jan. & May 1976 – NLO |
| X-705 U <sub>3</sub> O <sub>8</sub> – UF <sub>6</sub> Filter Ash | 2/57 – 7/77        | TRU's known to be processed Jan./Feb. 1974 – ICPP, and Jan. & May 1976 – NLO |
| GDP Cascade Operations   | Entire Period      | RU constituents concentrated at feed points                                  |
| GDP Cascade Operations   | Pre 1975           | Concentrated <sup>99</sup> Tc in purge cells                                 |
| X-326 MgF <sub>2</sub> <sup>99</sup> Tc Traps                    | After 1975         | Successfully removed <sup>99</sup> Tc in top of cascade                      |
| X-701B Holding Pond  | Before 1984        | Collected <sup>99</sup> Tc from X-705 solution recovery                      |
| X-705 Heavy Metals Precipitation                                 | 1984 and after     | Collects current <sup>99</sup> Tc from X-705 solution recovery               |
| <b>Site Specific Issues</b>                                      |                    |  |
| HEU refeed of RU-UF <sub>6</sub>                                 | 1/97 – 6/98        | 1.1 MTU French<br>0.3 MTU NUMEC  |
| Side Purge Fire  | 12/98              | Possible involvement with TRU materials                                      |

## 2.4 Activities where workers were likely to be in contact with recycled uranium through direct physical contact or airborne dust.

Table 2.4.1 summarizes activities where workers were likely to have been in contact with RU through direct physical contact or airborne dust at PORTS.

**Table 2.4-1****Activities at PORTS Where Workers Were Most Likely to Contact RU**

| Activity  | Comment   | TRU/FP |
|---|---|--------|
| Oxide Sampling (X-744G)   | Done in hood                                    | TRU    |
| Oxide Analysis (X-710)  | Generally done in hood                          | TRU    |
| Removal and disassembly of cascade equipment near TRU feed points       | In process building and X-705 high bay          | TRU    |
| Oxide Conversion Operation (X-705)                                      | High airborne uranium levels                    | TRU    |
| Oxide Conversion Maintenance (X-705)                                    | Filter handling for ash clean-out, ash handling | TRU    |
| MgF <sub>2</sub> and Alumina Trap Change-out and Media Handling (X-326) | Maybe HEU removal issue                         | FP     |
| X-701B Sludge Handling  | Pond sludge material to Envirocare              | FP     |
| X-705 Heavy Metals Precipitation Handling                               | Shipped to Envirocare                           | FP     |

**2.5 Activities that caused reportable environmental releases of recycled uranium constituents**

The only record of reported environmental releases of TRU elements occurred between October 1976 and March 1977. One sample above the minimum detection limit (MDL) was detected in the outfall from X-701B. A concentration of Np and Pu of  $3.7 \times 10^{-8}$  uCi/ml was measured which is just slightly above the MDL (at that time) of  $2.3 \times 10^{-8}$  uCi/ml. Activities ongoing on or before this time were recovery of uranium solutions and fluorination of oxides that later were discovered to have TRU constituents. Specific containers and sources of specific TRU constituents could not be accurately determined due to timing and cross flow anomalies. It is speculated that the source was the NLO oxides processed in Jan./May 1976.

Measurable and reported quantities of <sup>99</sup>Tc released to the environment have occurred since initial recognition of the concern as shown in Table 2.5-1.

As discussed earlier in this report, the activities resulting in surface water releases are those associated with recovery of uranium in X-705 and any episodes where solutions may have bypassed uranium recovery. Since uranium recovery processed materials generated from a myriad of internal plant sources (large and small parts cleaning, cylinder cleaning, field decontamination, etc.), the attribution to each source is somewhat speculative. The vast majority of these surface water releases are, however, felt to have originated from large parts (tunnel) operations where cascade equipment (especially converters) involved in CIP/CUP was processed.

Airborne releases of <sup>99</sup>Tc are felt to be predominantly attributable to cascade purging operations (top and side) when trap media became saturated or when trap media were ineffective due to extremely low concentrations of <sup>99</sup>Tc.

Environmental monitoring for TRU/FP appears to have been largely nonexistent prior to the mid 1970's. Beginning with the discovery of <sup>99</sup>Tc in the east drainage ditch (outfall 001) during the first quarter of FY 1975, sampling activities ramped up significantly. Since that discovery, and continuing until 1995, annual environmental reports have been issued that quantify <sup>99</sup>Tc releases and report any detection of TRU. Monitoring of air and water were and are conducted using both fixed sampling instruments and programmatic grab samples. Initially, all surface water sampling was manual. Current systems are capable of either time proportional or flow proportional sampling.

Current outfall sampling is done on a continuous basis, with seven day composites analyzed for gross alpha, beta, uranium, and  $^{99}\text{Tc}$ .

Air monitoring has taken a similar evolutionary course. As part of the original complement of equipment, ionization chamber instruments (space recorders) were installed on cascade vents for detecting  $\text{UF}_6$  releases. These later served to also detect the presence of  $^{99}\text{Tc}$  in-as-much as the equipment would become dysfunctional in the presence of  $^{99}\text{Tc}$  due to a rapid buildup in background radiation reading. A continuous sampling system installed during FY 1984 takes flow proportional samples and passes the gas through small calibrated alumina traps. Weekly (or more often if an anomaly occurs), the alumina is changed out and counted using radiochemistry techniques for uranium,  $^{235}\text{U}$ , and  $^{99}\text{Tc}$ . Data from this process takes as long as two weeks to receive, but is felt to be more accurate for quantifying releases.

**Table 2.5-1**

**Annual Releases of  $^{99}\text{Tc}$  to Surface Water and Air at PORTS**

| Fiscal Year  | Annual Releases of $^{99}\text{Tc}$ |             |            |                       |             |             |
|--------------|-------------------------------------|-------------|------------|-----------------------|-------------|-------------|
|              | To Surface Water (Ci)               | To Air (Ci) | Total (Ci) | To Surface Water (kg) | To Air (kg) | Total (kg)  |
| 1975         | 77.5                                |             | 77.5       | 4.5                   |             | 4.5         |
| 1976         | 19.1                                |             | 19.1       | 1.1                   |             | 1.1         |
| 1977         | 31.0                                | 4.5         | 35.5       | 1.819                 | 0.264       | 2.083       |
| 1978         | 17.7                                | 0.823       | 18.523     | 1.039                 | 0.048       | 1.087       |
| 1979         | 2.790                               | 0.170       | 2.96       | 0.164                 | 0.010       | 0.174       |
| 1980         | 7.740                               | 0.210       | 7.95       | 0.454                 | 0.012       | 0.467       |
| 1981         | 24.720                              | 0.108       | 24.828     | 1.451                 | 0.006       | 1.457       |
| 1982         | 11.840                              | 11.100      | 22.94      | 0.695                 | 0.651       | 1.346       |
| 1983         | 2.990                               | 0.561       | 3.551      | 0.175                 | 0.033       | 0.208       |
| 1984         | 9.340                               | 0.127       | 9.467      | 0.548                 | 0.007       | 0.556       |
| 1985         | 8.450                               | 0.123       | 8.573      | 0.496                 | 0.007       | 0.503       |
| 1986         | 2.480                               | 0.122       | 2.602      | 0.146                 | 0.007       | 0.153       |
| 1987         | 1.220                               | 0.169       | 1.389      | 0.072                 | 0.010       | 0.082       |
| 1988         | 0.870                               | 0.162       | 1.032      | 0.051                 | 0.010       | 0.061       |
| 1989         | 0.480                               | 0.096       | 0.576      | 0.028                 | 0.006       | 0.034       |
| 1990         | 1.350                               | 0.049       | 1.399      | 0.079                 | 0.003       | 0.082       |
| 1991         | 0.330                               | 0.044       | 0.374      | 0.019                 | 0.003       | 0.022       |
| 1992         | 0.210                               | 1.230       | 1.44       | 0.012                 | 0.072       | 0.085       |
| 1993         | 0.580                               | 7.830       | 8.41       | 0.034                 | 0.459       | 0.494       |
| 1994         | 0.167                               | 0.122       | 0.289      | 0.010                 | 0.007       | 0.017       |
| 1995         | --                                  | 0.0147      | 0.0147     |                       | 0.001       | 0.001       |
| <b>TOTAL</b> | <b>221</b>                          | <b>28</b>   | <b>249</b> | <b>12.98</b>          | <b>1.62</b> | <b>14.6</b> |